

02 APR 2001

## EXPRESS MAIL LABEL NUMBER EL619399881US

Form PTO-1390 REV.10-94)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEYS DOCKET NUMBER <b>RN98131</b>
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/elected office (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 1.6) <b>09/806629</b> NOTE: APPROPRIATE	
INTERNATIONAL APPLICATION NO. <b>PCT/FR99/02345</b>	INTERNATIONAL FILING DATE <b>October 1st, 1999</b>	PRIORITY DATE CLAIMED <b>October 2nd, 1998</b>	
<b>TITLE OF INVENTION</b> <b>DENTAL COMPOSITION BASED ON SILICONE CROSSLINKABLE BY CATION PROCESS</b>			
<b>APPLICANT (S) FOR DO/EO/US:</b> <b>Jean-Marc FRANCES</b>			
<p>Applicant herewith submits the United States Designated/Elected Office (DO/EO/US) the following items and other information:</p> <p>1 <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U S C 371      2 <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U S C 371      3 <input checked="" type="checkbox"/> This express request to begin national examination procedures (35 U S C 371) (f) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U S C 371 (b) and PCT Articles 22 and 39(1)      4 <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date      5 <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U S C 371(c)(2))          a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).          b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau.          c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US)      6 <input checked="" type="checkbox"/> A translation of the International Application into English (35 U S C 371(c)(2)).      7 <input checked="" type="checkbox"/> A copy of the International Search Report (PCT/ISA/210)      8 <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U S C 371(c)(2)).          a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).          b. <input type="checkbox"/> have been transmitted by the International Bureau.          c. <input type="checkbox"/> have not been made, however the time limit for making such amendments has NOT expired.          d. <input checked="" type="checkbox"/> have not been made and will not be made.      9 <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U S C 371(c)(3)).      10 <input checked="" type="checkbox"/> A oath or declaration of the inventor(s) (35 U S C 371(c)(4)). (<b>unsigned</b>)      11 <input checked="" type="checkbox"/> A copy of the International Preliminary Examination Report (PCT/IPEA/409).      12 <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U S C 371(c)(5)).  <b>Items 13 to 18 below concern document(s) or information included:</b>      13 <input checked="" type="checkbox"/> An information Disclosure Statement under 37 CFR 1.97 and 1.98      14 <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included      15 <input checked="" type="checkbox"/> A <b>FIRST</b> preliminary amendment      16 <input type="checkbox"/> A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment      17 <input type="checkbox"/> A substitute specification.      18 <input type="checkbox"/> A change of power of attorney and/or address letter      19 <input checked="" type="checkbox"/> Certificate of Mailing by Express Mail      20 <input checked="" type="checkbox"/> Other items or information   </p>			
<b>Forms PCT/IB/308 and 332, PCT/ISA/ 210 (English &amp; French Version), PCT/IPEA/ 409, PCT/FR99/02345 as published</b>			

US APPLICATION NO. <b>09/806629</b>	International Application No. <b>PCT/FR99/02345</b>	ATTORNEY DOCKET NUMBER <b>RN98131</b>																																																				
<b>CALCULATIONS PTO ONLY</b>																																																						
<p>21 The following fees are submitted:</p> <p><b>BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):</b></p> <p><input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor International search fee (37 CFR 1.445(a)(2) paid to USPTO and International Search Report not prepared by the EPO or JPO..... \$1,000.00</p> <p><input checked="" type="checkbox"/> International preliminary examination fee not paid to USPTO but International Search Report prepared by the EPO or JPO..... \$ 860.00</p> <p><input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search (37 CFR 1.4445 (a)(2)) fee paid to USPTO..... \$ 710.00</p> <p><input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... \$ 690.00</p> <p><input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4) ..... \$ 100.00</p> <p><b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b></p>																																																						
<p>Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).</p> <table border="1"> <tr> <td><b>CLAIMS</b></td> <td><b>NUMBER FILED</b></td> <td><b>NUMBER EXTRA</b></td> <td><b>RATE</b></td> </tr> <tr> <td>Total Claims</td> <td>8 -20=</td> <td>0</td> <td>X\$18.00</td> </tr> <tr> <td>Independent Claims</td> <td>1 -3=</td> <td>0</td> <td>X\$80.00</td> </tr> <tr> <td>MULTIPLE DEPENDENT CLAIMS(S) (if applicable)</td> <td>N/A</td> <td></td> <td>\$ 0.00</td> </tr> <tr> <td colspan="3"><b>TOTAL OF ABOVE CALCULATIONS =</b></td> <td>\$ 860.00</td> </tr> <tr> <td colspan="3">Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28)</td> <td>\$ 0.00</td> </tr> <tr> <td colspan="3"><b>SUBTOTAL =</b></td> <td>\$ 860.00</td> </tr> <tr> <td colspan="3">Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492 (f)).</td> <td>\$ 0.00</td> </tr> <tr> <td colspan="3"><b>TOTAL NATIONAL FEE =</b></td> <td>\$ 860.00</td> </tr> <tr> <td colspan="3">Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property + <input checked="" type="checkbox"/></td> <td>\$ 0.00</td> </tr> <tr> <td colspan="3"><b>TOTAL FEES ENCLOSED =</b></td> <td>\$ 860.00</td> </tr> <tr> <td colspan="3"></td> <td>Amount to be refunded \$</td> </tr> <tr> <td colspan="3"></td> <td>charged \$</td> </tr> </table>			<b>CLAIMS</b>	<b>NUMBER FILED</b>	<b>NUMBER EXTRA</b>	<b>RATE</b>	Total Claims	8 -20=	0	X\$18.00	Independent Claims	1 -3=	0	X\$80.00	MULTIPLE DEPENDENT CLAIMS(S) (if applicable)	N/A		\$ 0.00	<b>TOTAL OF ABOVE CALCULATIONS =</b>			\$ 860.00	Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28)			\$ 0.00	<b>SUBTOTAL =</b>			\$ 860.00	Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492 (f)).			\$ 0.00	<b>TOTAL NATIONAL FEE =</b>			\$ 860.00	Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property + <input checked="" type="checkbox"/>			\$ 0.00	<b>TOTAL FEES ENCLOSED =</b>			\$ 860.00				Amount to be refunded \$				charged \$
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<p>a <input type="checkbox"/> A check in the amount of \$_____ to cover the above fees is enclosed</p> <p>b <input checked="" type="checkbox"/> Please charge my Deposit Account No. <b>18-1171</b> in the amount of <b>\$860.00</b> to cover the above fees.</p> <p>c <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 18-1171 A duplicate copy of this sheet is enclosed.</p>																																																						
<p><b>NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.</b></p> <p><i>[Signature]</i></p> <p><b>SEND ALL CORRESPONDENCE TO:</b>  <b>Jean-Louis SEUGNET</b>  <b>RHODIA INC.</b>  <b>259 Prospect Plains Road</b>  <b>CN 7500</b>  <b>Cranbury, NJ 08512</b></p> <p><b>Jean-Louis SEUGNET</b>  <b>NAME</b>  <b>Limited Recognition under 37 CFR § 10.9(b)</b>  <b>enclosed.</b></p>																																																						
<p><b>April 02 , 2001</b></p> <p><b>DATE</b></p> <p><b>REGISTRATION NUMBER</b></p>																																																						

Case RN98131

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In the Application of: Jean-Marc FRANCES  
National Phase of PCT/FR99/02345  
International Filing Date : October 1st, 1999

Examiner: N/A

Art Unit: N/A

Serial No: N/A

Filing Date: April 02, 2001

For: DENTAL COMPOSITION BASED ON SILICONE CROSSLINKABLE  
BY CATION PROCESS

Assistant Commissioner for Patents  
Washington, D.C. 20231

PRELIMINARY AMENDMENT

Dear Sir:

Prior to calculation of filing fee, please enter the following amendment in the specification and claims:

In the specification:

Page 1, just after the title, please add: -- This application is an application under 35 U.S.C. Section 371 of International Application Number PCT/FR99/02345 filed on October 1st, 1999. --

In the claims:

Please cancel claims 1 to 11 and replace them with the following new claims 12 to 19:

12. (New) A dental composition comprising:

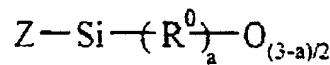
(1) at least one crosslinkable or polymerizable silicone oligomer or polymer which is liquid at room temperature or which is heat-meltable at a temperature of less than 100°C, and which comprises:

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a) at least one unit of formula (FS):



wherein:

a = 0, 1 or 2,

-  $\text{R}^0$ , identical or different, represents an alkyl, cycloalkyl, aryl,

vinyl, hydroxyl or alkoxy radical,

- Z, identical or different, is an organic substituent comprising at least one reactive epoxy, alkenyl ether, oxetane, dioxolane or carbonate functional group, and

b) at least two silicon atoms,

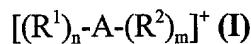
(2) at least one aromatic hydrocarbon photosensitizer with one or more aromatic nuclei which are optionally substituted, and having a residual light absorption of between 200 and 500 nm,

(3) at least one dental filler present in a proportion of at least 10% by weight relative to the total weight of the composition, and

(4) an effective quantity of at least one borate

photoinitiator having an anionic borate entity and a cationic borate entity and whose cationic entity is:

onium salts of formula (I):



in which formula:

A represents an element of groups 15 to 17 of valency v,

$R^1$  represents a carbocyclic or heterocyclic  $C_6\text{-}C_{20}$  aryl radical, optionally containing a nitrogen or sulfur atom,

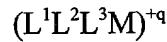
$R^2$  represents  $R^1$  or a linear or branched  $C_1\text{-}C_{30}$  alkyl or alkenyl radical; said radicals  $R^1$  and  $R^2$  being optionally substituted with a  $C_1\text{-}C_{25}$  alkoxy,

$C_1\text{-}C_{25}$  alkyl, nitro, chloro, bromo, cyano, carboxyl, ester or mercapto group,

n is an integer ranging from 1 to  $v + 1$ , v being the valency of the element A,

m is an integer ranging from 0 to  $v - 1$  with  $n + m = v + 1$ ,

(\*\*) organometallic salts of formula (III):



in which formula:

M represents a group 4 to 10 metal,

$L^1$  represents 1 ligand bound to the metal M by  $\pi$  by electrons, said ligand being  $\eta^3$ -alkyl,  $\eta^5$ -cyclopentadienyl,  $\eta^7$ -cycloheptatrienyl, optionally substituted  $\eta^6$ -aromatic compounds, or compounds having from 2 to 4 condensed rings, each ring being capable of contributing to the valency layer of the metal M by 3 to 8  $\pi$  electrons;

$L^2$  represents a ligand bound to the metal M by  $\pi$  electrons, said ligand being  $\eta^7$ -cycloheptatrienyl, optionally substituted  $\eta^6$ -benzene, or compounds having

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**Preliminary Amendment**

**PCT/FR99/02345**

from 2 to 4 condensed rings, each ring being capable of contributing to the valency layer of the metal M by 6 or 7  $\pi$  electrons;

$L^3$  represents from 0 to 3 ligands, which are identical or different, linked to the metal M by  $\sigma$  electrons, said ligand(s) being CO, or NO<sub>2</sub><sup>+</sup>; the total electron charge q of the complex to which  $L^1$ ,  $L^2$  and  $L^3$  contribute and the ionic charge of the metal M being positive and equal to 1 or 2; and

whose anionic borate entity is of the formula [BX<sub>a</sub>R<sub>b</sub>]<sup>-</sup> wherein:

- a and b are integers ranging, for a, from 0 to 3 and, for b, from 1 to 4 with a + b = 4,

- the symbols X represent:

a halogen atom (chlorine, fluorine) with a = 0 to 3,

an OH functional group with a = 0 to 2,

- the symbols R are identical or different and represent:

a phenyl radical substituted with at least one electron-attracting group or with at least 2 halogen atoms, when the cationic entity is an onium of an element of groups 15 to 17,

a phenyl radical substituted with at least one element or one electron-attracting group, when the cationic entity is an organometallic complex of an element of groups 4 to 10,

an aryl radical containing at least two aromatic nuclei optionally substituted with at least one electron-attracting group or element, regardless of the cationic entity.

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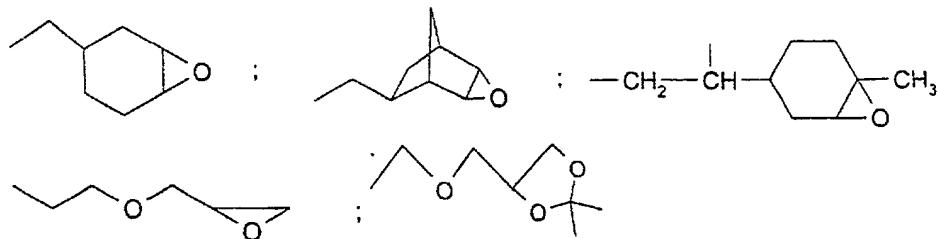
Preliminary Amendment

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13. (New) A composition as claimed in claim 12, wherein Z is an organic substituent Z1 comprising at least one reactive epoxy, or dioxolane functional group.

14. (New) A composition as claimed in claim 13, wherein the oligomer or polymer (1) further comprises other reactive functional groups Z which are alkenyl ether, oxetane or carbonate functional groups Z2.

15. (New) A composition as claimed in claim 12, wherein the reactive functional group(s) of Z1 are selected from the group consisting of the following radicals:



16. (New) A composition as claimed in claim 12, wherein the photoinitiator is

$[(\Phi_2 I)^+, [B(C_6F_5)_4]^-$	$[(C_8H_{17})-O-\Phi-I-\Phi)]^+, [B(C_6F_5)_4]^-$
$[C_{12}H_{25}-\Phi-I-\Phi]^+, [B(C_6F_5)_4]^-$	$[(C_8H_{17}-O-\Phi)_2I]^+, [B(C_6F_5)_4]^-$
$[(C_8H_{17})-O-\Phi-I-\Phi)]^+, [B(C_6F_5)_4]^-$	$[(\Phi)_3S]^+, [B(C_6F_5)_4]^-$
$[(\Phi)_2S-\Phi-O-C_8H_{17}]^+, [B(C_6H_4CF_3)_4]^-$	$[(C_{12}H_{25}-\Phi)_2I]^+, [B(C_6F_5)_4]^-$
$[(\Phi)_3S]^+, [B(C_6F_4OCF_3)_4]^-$	$[(\Phi-CH_3)_2I]^+, [B(C_6F_5)_4]^-$
$[(\Phi-CH_3)_2I]^+, [B(C_6F_4OCF_3)_4]^-$	$[CH_3-\Phi-I-\Phi-CH(CH_3)_2]^+, [B(C_6F_5)_4]^-$

$(\eta^5\text{-cyclopentadienyl})(\eta^6\text{-toluene}) Fe^+, [B(C_6F_5)_4]$

$(\eta^5\text{-cyclopentadienyl})(\eta^6\text{-methyl-1-naphthalene}) Fe^+, [B(C_6F_5)_4]$ , or

$(\eta^5\text{-cyclopentadienyl})(\eta^6\text{-cumene}) Fe^+, [B(C_6F_5)_4]$ .

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17. (New) A composition as claimed in claim 12, wherein the photosensitizer is:

4,4'-dimethoxybenzoin; 2-4-diethylthioxanthone

2-ethylanthraquinone; 2-methylanthraquinone;

1,8-dihydroxyanthraquinone; dibenzoylperoxide;

2,2-dimethoxy-2-phenylacetophenone;

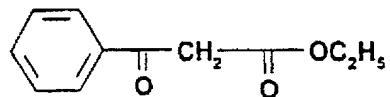
benzoin;

2-hydroxy-2-methylpropiophenone;

benzaldehyde;

4-(2-hydroxyethoxy)phenyl-(2-hydroxy-2-methylpropyl)-ketone;

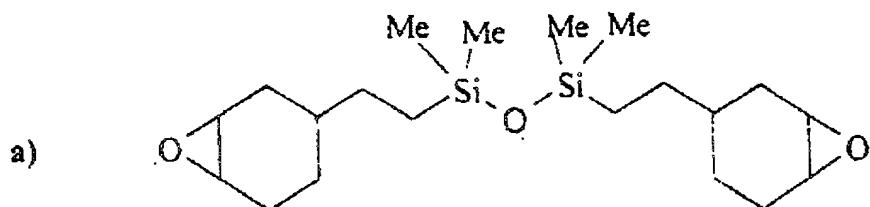
benzoylacetone;

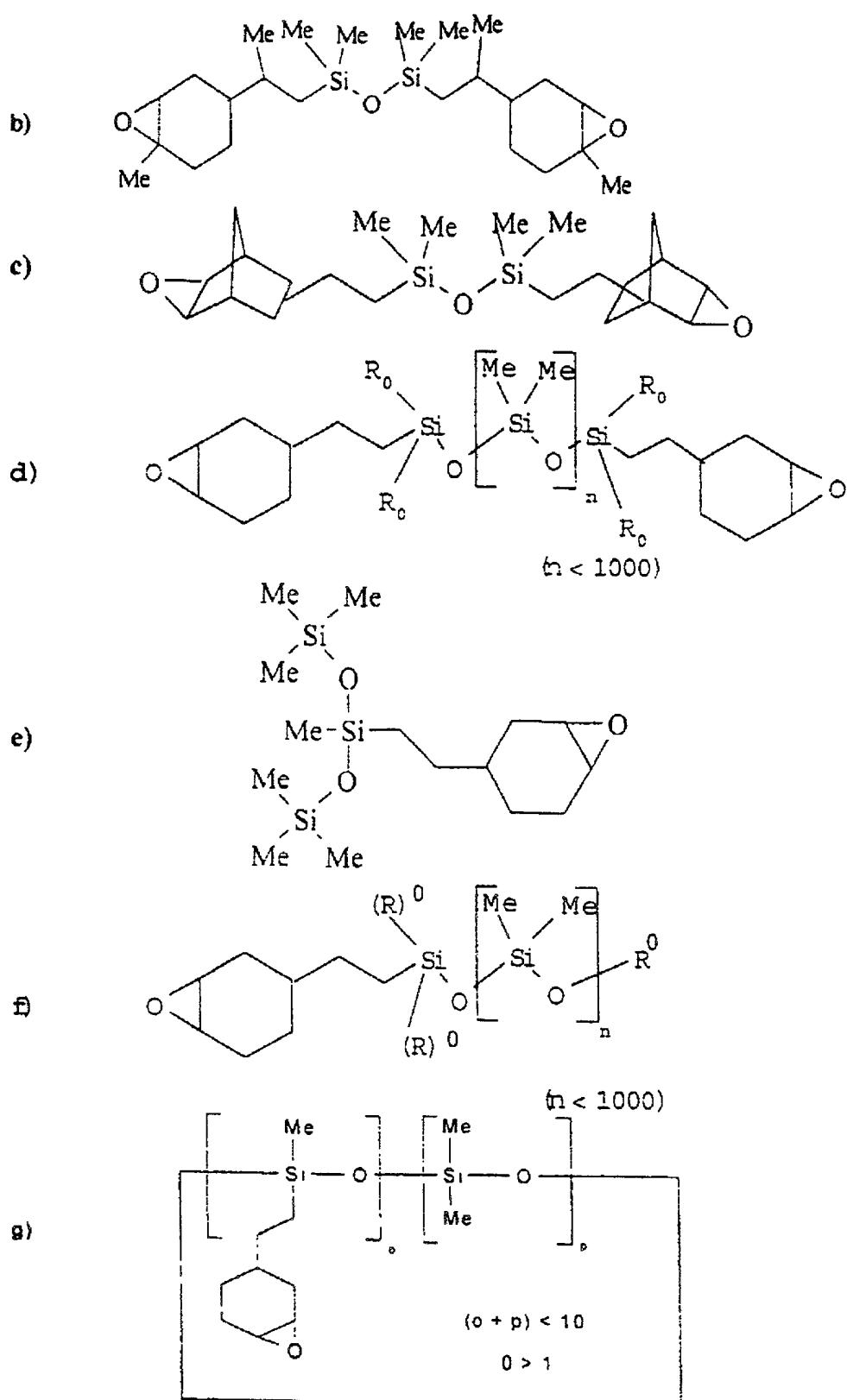


2-isopropylthioxanthone; 1-chloro-4-propoxythioxanthone; or

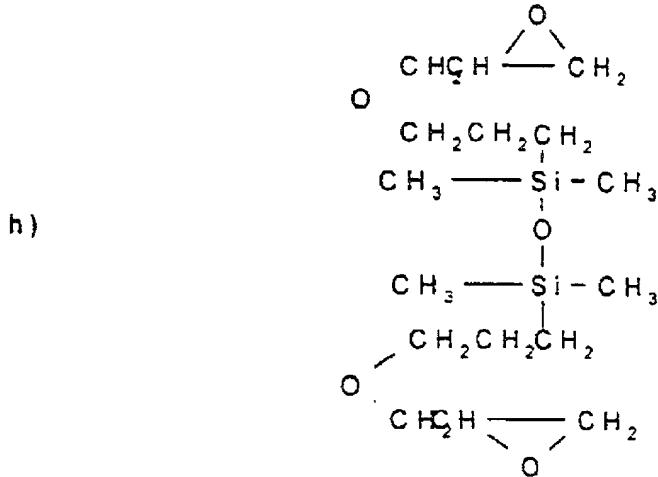
4-isopropylthioxanthone.

18. (New) A dental composition as claimed in claim 12, wherein the silicone oligomer or polymer (1) comprises at least one silicone having the following average formula:





or



19. (New) A process for the preparation of a dental prosthesis or dental restoration, comprising the step of using a dental composition as defined in claim 12.

#### REMARKS

It is asserted that these amendments do not add new matter. Support for these amendments can be found in the specification and claims as originally filed. Entry of these amendments is respectfully requested.

Respectfully submitted,

By

April 01, 2001

JEAN-LOUIS SEUGNET

Limited Recognition under 37 CFR § 10.9(b)  
enclosed

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CN7500,  
Cranbury, NJ 08512

EL619399881US

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PCT/FR99/02345

DENTAL COMPOSITION BASED ON SILICONE CROSSLINKABLE BY  
CATION PROCESS

The field of the invention is that of dental compositions. More precisely, the dental compositions used in the context of the present invention can be used for producing dental prostheses and for dental restoration.

Up until now, to produce dental compositions for the preparation of dental prostheses or of dental restoration materials, it is possible to use resins based on photopolymerizable acrylates. These ready-to-formulate products exhibit however upon use problems of irritation and potential problems of toxicity.

In addition, these products have the major disadvantage of causing high volume shrinkage during their polymerization, which makes their use complex and difficult for the production of dental prostheses or of dental restoration materials. Problems of attachment due to the volume shrinkage or to the lack of adherence of the polymers used are in particular observed.

The object of the present invention is to provide novel dental compositions which do not exhibit the disadvantages of the prior art. These novel dental compositions, which are polymerizable and/or crosslinkable in an oral environment, have markedly improved qualities, in particular as regards the very marked reduction in the phenomenon of shrinkage of the

dental compositions used for the production of dental prostheses or of dental restoration materials.

The polymerizable and/or crosslinkable dental composition according to the invention comprises:

- 5                     (1) at least one crosslinkable and/or polymerizable silicone oligomer or polymer which is liquid at room temperature or which is heat-meltable at a temperature of less than 100°C, and which comprises:
- 10                     • at least one unit of formula (FS):  
$$Z-\text{Si}-(\overset{\overset{R^0}{|}}{a})-\text{O}_{(3-a)/2}$$
in which:  
- a = 0, 1 or 2,  
- R<sup>0</sup>, identical or different, represents an alkyl, cycloalkyl, aryl, vinyl, hydroxyl or alkoxy radical, preferably a C<sub>1</sub>-C<sub>6</sub> lower alkyl,  
- Z, identical or different, is an organic substituent comprising at least one reactive epoxy, and/or alkenyl ether and/or oxetane and/or dioxolane and/or carbonate functional group, and preferably Z being an organic substituent comprising at least one reactive epoxy and/or dioxolane functional group,  
• and at least two silicon atoms,
- 15  
20  
25

(2) an effective quantity of at least one borate-type photoinitiator,

5 (3) at least one aromatic hydrocarbon photosensitizer with one or more aromatic nuclei which are substituted or not, having a residual light absorption of between 200 and 500 nm,

10 (4) and at least one dental filler present in a proportion of at least 10% by weight relative to the total weight of the composition.

According to a first advantageous variant of the present invention, the dental composition is polymerizable and/or crosslinkable under activation by 15 the thermal route and/or by the photochemical route.

In general, the photochemical activation is carried out under UV radiation. More particularly, UV radiation having a wavelength of the order of 200 to 500 nm is used for the production of dental prostheses 20 and UV-visible radiation having a wavelength greater than 400 nm for the production of restoration materials. A wavelength greater than 400 nm allows crosslinking and/or polymerization in an oral environment.

25 The silicone polymer or oligomer (1) has the advantage, compared with organic resins which are crosslinked by the cationic route, of being transparent

to UV-visible light and therefore its use makes it possible to obtain materials which are very thick and whose photocrosslinking occurs in a short time.

The reactive functional groups Z of the  
5 silicone polymer or oligomer (1) may be highly varied. However, particularly advantageous dental compositions are obtained when the silicone oligomer or polymer (1) comprises at least one (FS) unit in which Z represents an organic substituent Z1 comprising at least one  
10 reactive epoxy, and/or dioxolane functional group, and preferably at least one reactive epoxy functional group.  
15

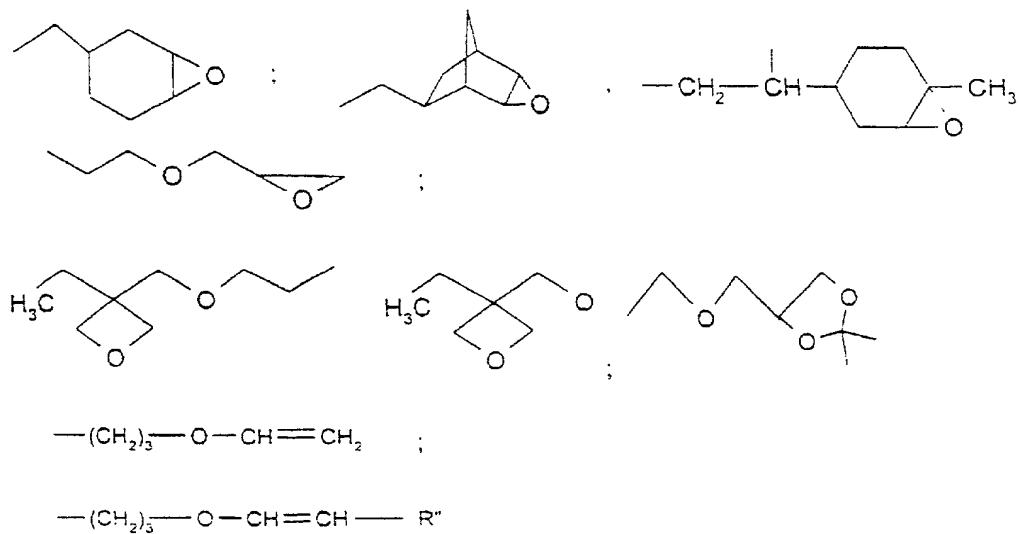
According to two advantageous alternatives of the present invention, the silicone oligomer or polymer  
15 (1) with at least one reactive epoxy and/or dioxolane functional group Z1, and preferably at least one reactive epoxy functional group may:

- (i) either comprise only this (these) type(s) of reactive functional group(s) Z1,  
20
- (ii) or comprise other reactive functional groups Z such as the reactive alkenyl ether, oxetane and/or carbonate functional groups Z2.

In the case of the first alternative (i), the  
25 dental composition may also comprise other silicone oligomers and/or polymers comprising other reactive functional groups Z2 such as alkenyl ether, oxetane

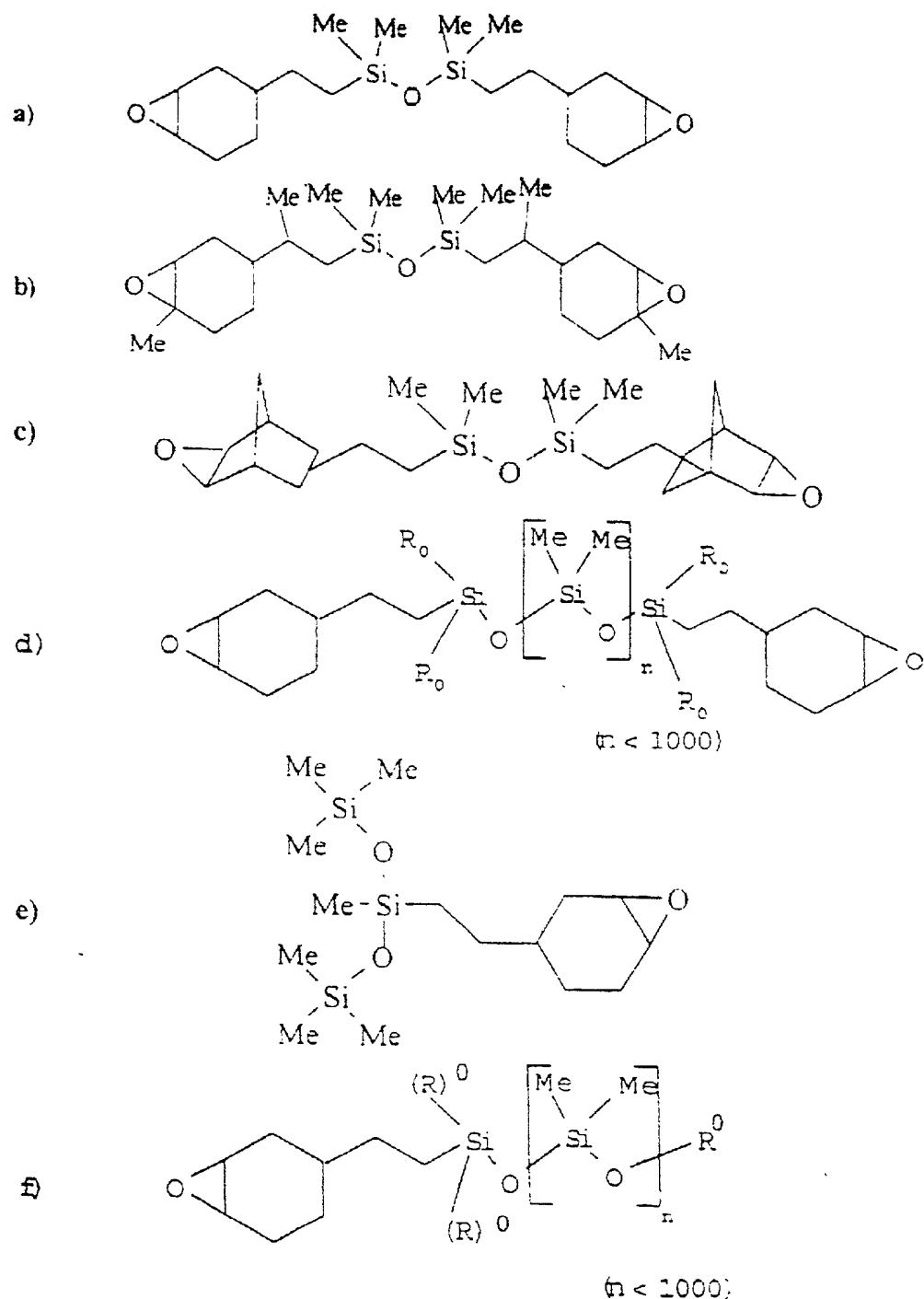
and/or carbonate functional groups and optionally reactive functional groups Z1.

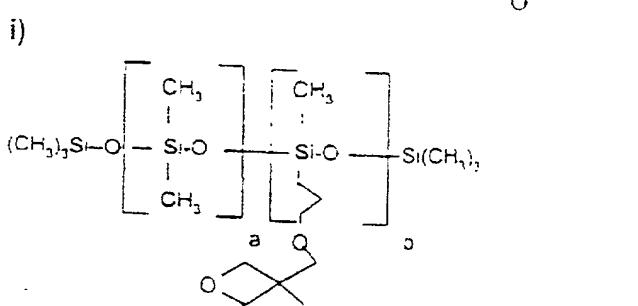
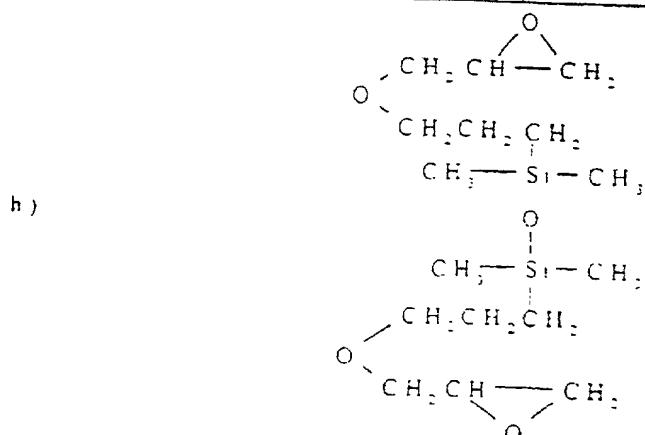
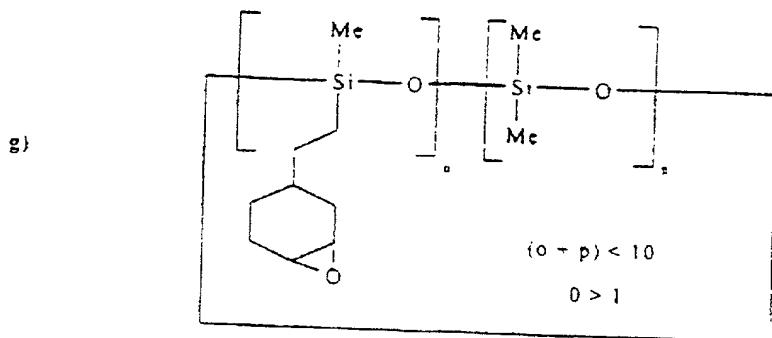
By way of examples of reactive functional groups Z, these may in particular be chosen from the 5 following radicals:



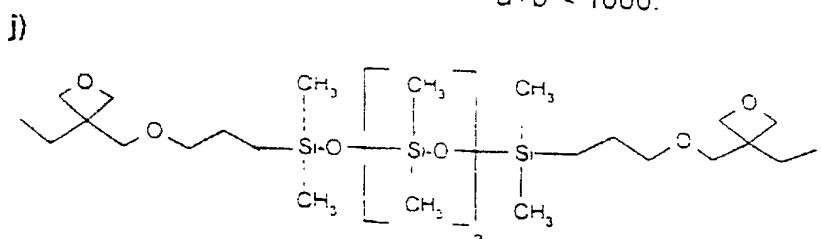
- with R" representing a linear or branched C<sub>1</sub>-C<sub>6</sub> alkyl radical.

According to a second advantageous variant of 10 the present invention, the silicone polymer or oligomer consists of at least one silicone having the following average formula:

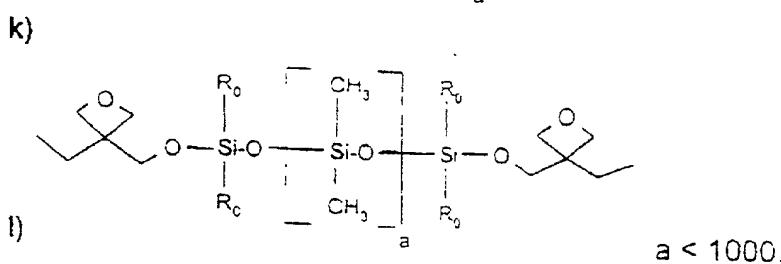




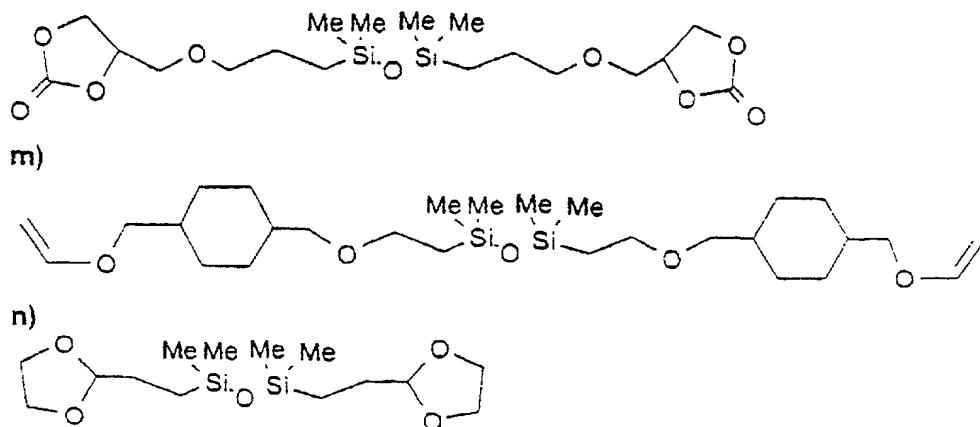
$a+b < 1000.$



$a < 1000.$



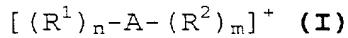
$a < 1000.$



The cationic photoinitiators are chosen from  
onium borates (taken on their own or as a mixture with  
each other) of an element of groups 15 to 17 of the  
5 Periodic Table [Chem. & Eng. News, vol. 63, No. 5, 26  
of 4 February 1985] or of an organometallic complex of  
an element of groups 4 to 10 of the Periodic Table  
[same reference].

The cationic entity of the borate is selected  
10 from:

(1) the onium salts of formula (I)



in which formula:

- A represents an element of groups 15 to 17 such  
as for example: I, S, Se, P or N,
- R<sup>1</sup> represents a carbocyclic or heterocyclic C<sub>6</sub>-C<sub>20</sub>  
aryl radical, it being possible for said  
heterocyclic radical to contain, as  
heteroelements, nitrogen or sulfur,

• R<sup>2</sup> represents R<sup>1</sup> or a linear or branched C<sub>1</sub>-C<sub>30</sub> alkyl or alkenyl radical; said radicals R<sup>1</sup> and R<sup>2</sup> being optionally substituted with a C<sub>1</sub>-C<sub>25</sub> alkoxy, C<sub>1</sub>-C<sub>25</sub> alkyl, nitro, chloro, bromo, cyano, carboxyl, ester or mercapto group,

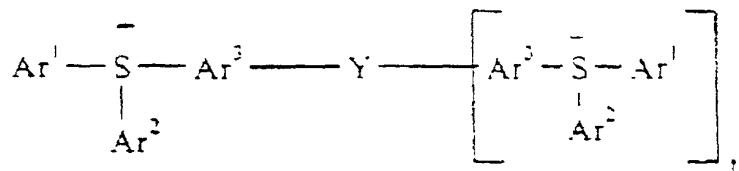
5           • n is an integer ranging from 1 to v + 1, v being the valency of the element A,

• m is an integer ranging from 0 to v - 1 with n + m = v + 1,

10           (2) the oxoisothiocromanium salts described in patent application WO 90/11303, in particular the sulfonium salt of 2-ethyl-4-oxoisothiocromanium or 2-dodecyl-4-oxoisothiocromanium,

15           (3) the sulfonium salts in which the cationic entity comprises:

• 3<sub>1</sub> at least one polysulfonium entity of formula (III.1):



in which:

20           - the symbols Ar<sup>1</sup>, which are identical or different, each represent a monovalent phenyl or naphthyl radical, optionally substituted with one or more radicals chosen from: a linear or branched C<sub>1</sub>-C<sub>12</sub>, preferably C<sub>1</sub>-C<sub>6</sub>, alkyl radical, a linear or branched C<sub>1</sub>-C<sub>12</sub>, preferably

C<sub>1</sub>-C<sub>6</sub>, alkoxy radical, a halogen atom, an -OH group, a -COOH group, an ester group -COO-alkyl where the alkyl portion is a linear or branched C<sub>1</sub>-C<sub>12</sub>, preferably C<sub>1</sub>-C<sub>6</sub>, residue, and a group of formula -Y<sup>4</sup>-Ar<sup>2</sup> where the symbols Y<sup>4</sup> and Ar<sup>2</sup> have the meanings given just below,

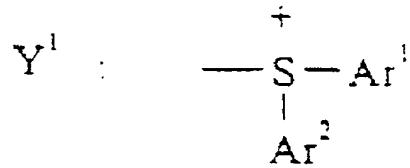
- the symbols Ar<sup>2</sup>, which are identical or different, each represent a monovalent phenyl or naphthyl radical, optionally substituted with one or more radicals chosen from: a linear or branched C<sub>1</sub>-C<sub>12</sub>, preferably C<sub>1</sub>-C<sub>6</sub>,

alkyl radical, a linear or branched C<sub>1</sub>-C<sub>12</sub>, preferably C<sub>1</sub>-C<sub>6</sub>, alkoxy radical, a halogen atom, an -OH group, a -COOH group, an ester group -COO-alkyl where the alkyl portion is a linear or branched C<sub>1</sub>-C<sub>12</sub>, preferably C<sub>1</sub>-C<sub>6</sub>, residue,

- the symbols Ar<sup>3</sup>, which are identical or different, each represent a divalent phenylene or naphthylene radical, optionally substituted with one or more radicals chosen from: a linear or branched C<sub>1</sub>-C<sub>12</sub>, preferably C<sub>1</sub>-C<sub>6</sub>, alkyl radical, a linear or branched C<sub>1</sub>-C<sub>12</sub>, preferably C<sub>1</sub>-C<sub>6</sub>, alkoxy radical, a halogen atom, an -OH group, a -COOH group, an ester group -COO-alkyl where the alkyl portion is a linear or branched C<sub>1</sub>-C<sub>12</sub>, preferably C<sub>1</sub>-C<sub>6</sub>, residue,

- t is an integer equal to 0 or 1, with the additional conditions according to which:

+ when t = 0, the symbol Y is then a monovalent radical Y<sup>1</sup> representing the group of formula:

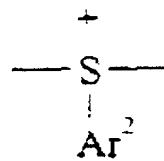


where the symbols  $Ar^1$  and  $Ar^2$  possess the meanings given above,

5      + when  $t = 1$ :

• on the one hand, the symbol  $Y$  is then a divalent radical having the following meanings  $Y^2$  to  $Y^4$ :

- $Y^2$ : a group of formula:



10     where the symbol  $Ar^2$  has the meanings given above,

- $Y^3$ : a single valency bond,
- $Y^4$ : a divalent residue chosen from:



a linear or branched  $C_1-C_{12}$ , preferably  $C_1-C_6$ ,  
15     alkylene residue and a residue of formula  
 $-Si(CH_3)_2O-$ ,

• on the other hand, in the case solely where the symbol  $Y$  represents  $Y^3$  or  $Y^4$ , the (terminal) radicals  $Ar^1$  and  $Ar^2$  possess, in addition to the meanings given above, the possibility of being linked to each other by the residue  $Y'$  consisting of  $Y'^1$  a single valency bond or of  $Y'^2$  a divalent

residue chosen from the residues cited in relation to the definition of Y<sup>4</sup>, which is inserted between the carbon atoms, opposite each other, situated on each aromatic ring at the ortho position with respect to the carbon atom directly linked to the cation S<sup>+</sup>;

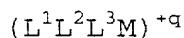
- 3<sub>2</sub> and/or at least one monosulfonium entity possessing a single cationic center S<sup>+</sup> per mol of cation and consisting, in most cases, of entity of formula (II.2):



10

in which Ar<sup>1</sup> and Ar<sup>2</sup> have the meanings given above in relation to formula (III.1), including the possibility of directly linking to each other only one of the radicals Ar<sup>1</sup> to Ar<sup>2</sup> in the manner indicated above in relation to the definition of the additional condition in force when t = 1 in formula (II), calling into play the residue Y' ;

(4) the organometallic salts of formula (III):



20 in which formula:

- M represents a group 4 to 10 metal, in particular iron, manganese, chromium or cobalt,
- L<sup>1</sup> represents 1 ligand bound to the metal M by  $\pi$  electrons, which ligand is chosen from the ligands 25  $\eta^3$ -alkyl,  $\eta^5$ -cyclopentadienyl and  $\eta^7$ -cycloheptatrienyl

and the  $\eta^6$ -aromatic compounds chosen from the optionally substituted  $\eta^6$ -benzene ligands and the compounds having from 2 to 4 condensed rings, each ring being capable of contributing to the valency layer of 5 the metal M by 3 to 8  $\pi$  electrons;

•  $L^2$  represents a ligand bound to the metal M by  $\pi$  electrons, which ligand is chosen from the ligands  $\eta^7$ -cycloheptatrienyl and the  $\eta^6$ -aromatic compounds chosen from the optionally substituted ligands 10  $\eta^6$ -benzene and the compounds having from 2 to 4 condensed rings, each ring being capable of contributing to the valency layer of the metal M by 6 or 7  $\pi$  electrons;

•  $L^3$  represents from 0 to 3 ligands, which are identical 15 or different, linked to the metal M by  $\sigma$  electrons, which ligand(s) is (are) chosen from CO and  $NO_2^-$ ; the total electron charge q of the complex to which  $L^1$ ,  $L^2$  and  $L^3$  contribute and the ionic charge of the metal M being positive and equal to 1 or 2;

20 The anionic borate entity has the formula  $[BX_aR_b]^-$  in which:

- a and b are integers ranging, for a, from 0 to 3 and, for b, from 1 to 4 with  $a + b = 4$ ,
- the symbols X represent:

25 \* a halogen atom (chlorine, fluorine) with a = 0 to 3,

\* an OH functional group with a = 0 to 2,

- the symbols R are identical or different and represent:

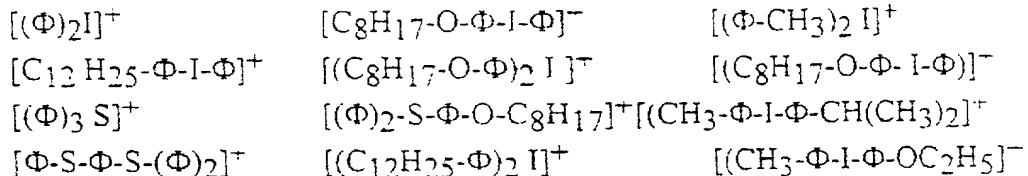
- a phenyl radical substituted with at least one electron-attracting group such as for example OCF<sub>3</sub>, CF<sub>3</sub>, NO<sub>2</sub>, CN, and/or with at least 2 halogen atoms (fluorine most particularly), this being when the cationic entity is an onium of an element of groups 15 to 17,
- a phenyl radical substituted with at least one element or one electron-attracting group, in particular a halogen atom (fluorine most particularly), CF<sub>3</sub>, OCF<sub>3</sub>, NO<sub>2</sub>, CN, this being when the cationic entity is an organometallic complex of an element of groups 4 to 10,
- an aryl radical containing at least two aromatic nuclei such as for example biphenyl, naphthyl, optionally substituted with at least one electron-attracting group or element, in particular a halogen atom (fluorine most particularly), OCF<sub>3</sub>, CF<sub>3</sub>, NO<sub>2</sub>, CN, regardless of the cationic entity.

Without being limiting, more details are given below as regards the subclasses of onium borate and of borate of organometallic salts more particularly preferred in the context of the use in accordance with the invention.

According to a first preferred variant of the invention, the entity of the anionic borate entity which are most particularly suitable are the following:

- |   |  |
|---|--|
| 1': [B(C <sub>6</sub> F <sub>5</sub> ) <sub>4</sub> ] <sup>-</sup>                    | 5': [B(C <sub>6</sub> H <sub>3</sub> (CF <sub>3</sub> ) <sub>2</sub> ) <sub>4</sub> ] <sup>-</sup> |
| 2': [(C <sub>6</sub> F <sub>5</sub> ) <sub>2</sub> BF <sub>2</sub> ] <sup>-</sup>     | 6': [B(C <sub>6</sub> H <sub>3</sub> F <sub>2</sub> ) <sub>4</sub> ] <sup>-</sup>                  |
| 3': [B(C <sub>6</sub> H <sub>4</sub> CF <sub>3</sub> ) <sub>4</sub> ] <sup>-</sup>    | 7': [C <sub>6</sub> F <sub>5</sub> BF <sub>3</sub> ] <sup>-</sup>                                  |
| 4': [B(C <sub>6</sub> F <sub>4</sub> OCF <sub>3</sub> ) <sub>4</sub> ] <sup>-</sup> . |  |

5 According to a second preferred variant of the invention, the onium salts (1) which can be used are described in numerous documents, in particular in patents US-A-4 026 705, US-A-4 032 673, US-A-4 069 056, US-A-4 136 102, US-A-4 173 476. Among these, the  
10 following cations will be most particularly preferred:



According to a third preferred variant, the organometallic salts (4) which can be used are  
15 described in the documents US-A-4 973 722, US-A-4 992 572, EP-A-203 829, EP-A-323 584 and EP-A-354 181. The organometallic salts most readily selected according to the invention are in particular:

- . (η<sup>5</sup>-cyclopentadienyl) (η<sup>6</sup>-toluene) Fe<sup>+</sup>,  
20 . (η<sup>5</sup>-cyclopentadienyl) (η<sup>6</sup>-methyl-1-naphthalene) Fe<sup>+</sup>,

- . ( $\eta^5$ -cyclopentadienyl) ( $\eta^6$ -cumene)  $\text{Fe}^+$ ,
- . bis( $\eta^6$ -mesitylene)  $\text{Fe}^+$ ,
- . bis( $\eta^6$ -benzene)  $\text{Cr}^+$ .

In agreement with these three preferred variants, the following products may be mentioned by way of examples of photoinitiators of the onium borate type:

- |  |   |
|--|---|
| $[(\Phi)_2 \text{I}]^+, [\text{B}(\text{C}_6\text{F}_5)_4]^-$  | $[(\text{C}_8\text{H}_{17})\text{-O-}\Phi\text{-I-}\Phi]^+, [\text{B}(\text{C}_6\text{F}_5)_4]^-$   |
| $[\text{C}_{12}\text{H}_{25}\text{-}\Phi\text{-I-}\Phi]^+, [\text{B}(\text{C}_6\text{F}_5)_4]^-$       | $[(\text{C}_8\text{H}_{17}\text{-O-}\Phi)_2\text{I}]^+, [\text{B}(\text{C}_6\text{F}_5)_4]^-$       |
| $[(\text{C}_8\text{H}_{17})\text{-O-}\Phi\text{-I-}\Phi]^+, [\text{B}(\text{C}_6\text{F}_5)_4]^-$      | $[(\Phi)_3\text{S}]^+, [\text{B}(\text{C}_6\text{F}_5)_4]^-$  |
| $[(\Phi)_2\text{S-}\Phi\text{-O-C}_8\text{H}_{17}]^+, [\text{B}(\text{C}_6\text{H}_4\text{CF}_3)_4]^-$ | $[(\text{C}_{12}\text{H}_{25}\text{-}\Phi)_2\text{I}]^+, [\text{B}(\text{C}_6\text{F}_5)_4]^-$      |
| $[(\Phi)_3\text{S}]^+, [\text{B}(\text{C}_6\text{F}_4\text{OCF}_3)_4]^-$                               | $[(\Phi\text{-CH}_3)_2\text{I}]^+, [\text{B}(\text{C}_6\text{F}_5)_4]^-$                            |
| $[(\Phi\text{-CH}_3)_2\text{I}]^+, [\text{B}(\text{C}_6\text{F}_4\text{OCF}_3)_4]^-$                   | $[\text{CH}_3\text{-}\Phi\text{-I-}\Phi\text{-CH(CH}_3)_2]^+, [\text{B}(\text{C}_6\text{F}_5)_4]^-$ |
- . ( $\eta^5$ -cyclopentadienyl) ( $\eta^6$ -toluene)  $\text{Fe}^+$ ,  $[\text{B}(\text{C}_6\text{F}_5)_4]^-$
- . ( $\eta^5$ -cyclopentadienyl) ( $\eta^6$ -methyl-1-naphthalene)  $\text{Fe}^+$ ,  $[\text{B}(\text{C}_6\text{F}_5)_4]^-$
- . ( $\eta^5$ -cyclopentadienyl) ( $\eta^6$ -cumene)  $\text{Fe}^+$ ,  $[\text{B}(\text{C}_6\text{F}_5)_4]^-$

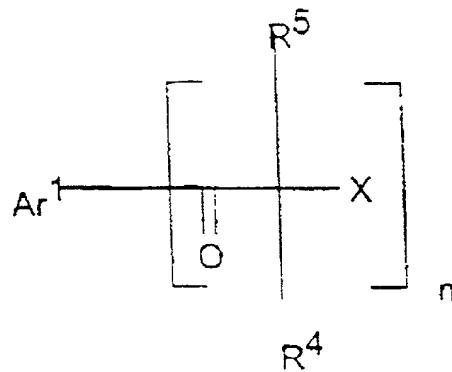
As another literary reference for defining the onium borates (1) and (2) and the borates of organometallic salts (4), there may be mentioned the entire content of patent applications EP 0 562 897 and 0 562 922. This content is integrally incorporated by reference into the present disclosure.

As another example of onium salt which can be used as photoinitiator, there may be mentioned those disclosed in American patents US 4 138 255 and US 4 310 469.

Other cationic photoinitiators may also be used, e.g.:

- those marketed by Union-Carbide (photoinitiator 6990 and 6974 triarylsulfonium hexafluorophosphate and hexafluoroantimonate),
- the salts of iodonium hexafluorophosphate or hexafluoroantimonate,
- or the ferrocenium salts of these various anions.

The nature of the photosensitizer contained in the dental composition according to the invention may be highly varied. In the context of the invention, it corresponds in particular to one of the following formulae (IV) to (XXII):



• formula (IV)

in which:

- when  $n = 1$ ,  $Ar^1$  represents an aryl radical containing from 6 to 18 carbon atoms, a

tetrahydronaphthyl, thietyl, pyridyl or furyl radical or a phenyl radical carrying one or more substituents chosen from the group consisting of F, Cl, Br, CN, OH, linear or branched C<sub>1</sub>-C<sub>12</sub> alkyls, -CF<sup>3</sup>, -OR<sup>6</sup>, -OPhenyl, -SR<sup>6</sup>, -SPhenyl, -SO<sub>2</sub>Phenyl, -COOR<sup>6</sup>, -O-(CH<sub>2</sub>-CH=CH<sub>2</sub>), -O(CH<sub>2</sub>H<sub>4</sub>-O)<sub>m</sub>-H, -O(C<sub>3</sub>H<sub>6</sub>O)<sub>m</sub>-H, m being between 1 and 100,

5 - when n = 2, **Ar<sub>1</sub>** represents a C<sub>6</sub>-C<sub>12</sub> arylene radical or a phenylene-T-phenylene radical where T represents -O-, -S-, -SO<sub>2</sub>- or -CH<sub>2</sub>-,

10 - **X** represents a group -OR<sup>7</sup> or -OSiR<sup>8</sup>(R<sup>9</sup>)<sub>2</sub> or forms, with R<sup>4</sup>, a group -O-CH(R<sup>10</sup>)-,

15 - **R<sub>4</sub>** represents a linear or branched C<sub>1</sub>-C<sub>8</sub> alkyl radical which is unsubstituted or which carries an -OH, -OR<sup>6</sup>, C<sub>2</sub>-C<sub>8</sub> acyloxy, -CF<sup>3</sup> or -CN group, a C<sub>3</sub> or C<sub>4</sub> alkenyl radical, a C<sub>6</sub> to C<sub>18</sub> aryl radical, a C<sub>7</sub> to C<sub>9</sub> phenylalkyl radical,

20 - **R<sup>5</sup>** has one of the meanings given for R<sup>4</sup> or represents a radical -CH<sub>2</sub>CH<sub>2</sub>R<sup>11</sup>, or alternatively forms with R<sup>4</sup> a C<sub>2</sub>-C<sub>8</sub> alkylene radical or a C<sub>3</sub>-C<sub>9</sub> oxa-alkylene or aza-alkylene radical,

25 - **R<sup>6</sup>** represents a lower alkyl radical containing from 1 to 12 carbon atoms,

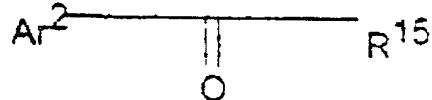
- **R<sup>7</sup>** represents a hydrogen atom, a C<sub>1</sub>-C<sub>12</sub> alkyl radical, a C<sub>2</sub>-C<sub>6</sub> alkyl radical carrying an -OH, -OR<sup>6</sup> or -CN group, a C<sub>3</sub>-C<sub>6</sub> alkenyl radical, a cyclohexyl or benzyl radical, a phenyl radical

optionally substituted with a chlorine atom or a linear or branched C<sub>1</sub>-C<sub>12</sub> alkyl radical, or a 2-tetrahydropyranyl radical,

5       - R<sup>8</sup> and R<sup>9</sup> are identical or different and each represent a C<sub>1</sub>-C<sub>4</sub> alkyl radical or a phenyl radical,

10      - R<sup>10</sup> represents a hydrogen atom, a C<sub>1</sub>-C<sub>8</sub> alkyl radical or a phenyl radical,

15      - R<sup>11</sup> represents a radical -CONH<sub>2</sub>, -CONHR<sup>6</sup>, -CON(R<sup>6</sup>)<sub>2</sub>, -P(O)(OR<sup>6</sup>)<sub>2</sub> or 2-pyridyl;



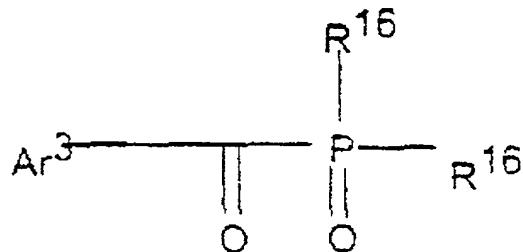
• formula (V)

in which:

15      - Ar<sup>2</sup> has the same meaning as Ar<sup>1</sup> of formula (IV)  
in the case where n = 1,

20      - R<sup>15</sup> represents a radical chosen from the group consisting of a radical Ar<sup>2</sup>, a linear or branched C<sub>1</sub>-C<sub>12</sub> alkyl radical, a C<sub>6</sub>-C<sub>12</sub> cycloalkyl radical, and a cycloalkyl radical forming a C<sub>6</sub>-C<sub>12</sub> ring with the carbon of the ketone or a carbon of the radical Ar<sup>2</sup>, it being possible for these radicals to be substituted with one or more substituents chosen from the group consisting of -F, -Cl, -Br, -CN, -OH, -CF<sub>3</sub>, -OR<sup>6</sup>, -SR<sup>6</sup>, -COOR<sup>6</sup>, the linear or branched C<sub>1</sub>-C<sub>12</sub> alkyl radicals optionally carrying

an -OH, -OR<sup>6</sup> and/or -CN group, and the linear or branched C<sub>1</sub>-C<sub>8</sub> alkenyl radicals;

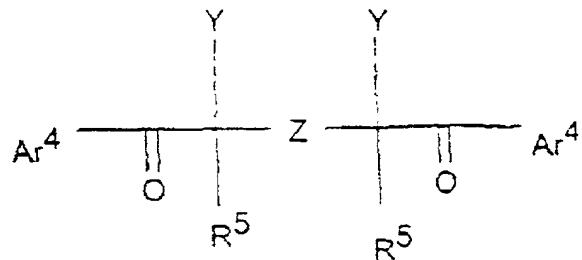


• formula (VI)

5        in which:

Ar<sup>3</sup> has the same meaning as Ar<sup>1</sup> of formula (IV) in the case where n = 1,

10        - R<sup>16</sup>, identical or different, represents a radical chosen from the group consisting of a radical Ar<sup>3</sup>, a radical -(C=O)-Ar<sup>3</sup>, a linear or branched C<sub>1</sub>-C<sub>12</sub> alkyl radical, a C<sub>6</sub>-C<sub>12</sub> cycloalkyl radical, it being possible for these radicals to be substituted with one or more substituents chosen from the group consisting of -F, -Cl, -Br, -CN, -OH, -CF<sub>3</sub>, -OR<sup>6</sup>, -SR<sup>6</sup>, -COOR<sup>6</sup>, the linear or branched C<sub>1</sub>-C<sub>12</sub> alkyl radicals optionally carrying an -OH, -OR<sup>6</sup> and/or -CN group, and the linear or branched C<sub>1</sub>-C<sub>8</sub> alkenyl radicals;



• formula (VII)

in which:

- **R<sup>5</sup>**, which are identical or different, have the same meanings as in formula (III),

5 - **y**, which are identical or different, represent X and/or R<sup>4</sup>,

- **z** represents:

• a direct bond,

10 • a C<sub>1</sub>-C<sub>6</sub> divalent alkylene radical, or a phenylene, diphenylene or phenylene-T-phenylene radical, or alternatively forms, with the two substituents R<sup>5</sup> and the two carbon atoms carrying these substituents, a cyclopentane or cyclohexane nucleus,

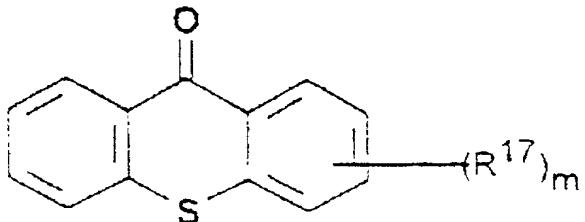
15 • a divalent group -O-R<sup>12</sup>-O-, -O-SiR<sup>8</sup>R<sup>9</sup>-O-SiR<sup>8</sup>R<sup>9</sup>-O-, or -O-SiR<sup>8</sup>R<sup>9</sup>-O-,

- **R<sup>12</sup>** represents a C<sub>2</sub>-C<sub>8</sub> alkylene, C<sub>4</sub>-C<sub>6</sub> alkenylene or xylylene radical,

and **Ar<sup>4</sup>** has the same meaning as Ar<sup>1</sup> of formula (IV)

20 in the case where n = 1.

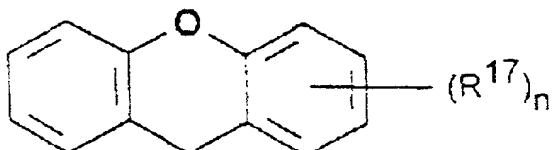
• family of thioxanthones of formula (VIII):



- m = 0 to 8,

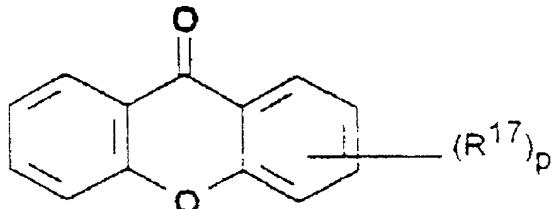
- R<sup>17</sup>, identical or different substituent(s) on the aromatic nucleus (nuclei), represent a linear or branched C1-C12 alkyl radical, a C6-C12 cycloalkyl radical, a radical Ar<sup>1</sup>, a halogen atom, an -OH, 5 -CN, -NO<sub>2</sub>, -COOR<sup>6</sup>, -CHO, Ophenyl, -CF<sub>3</sub>, -SR<sup>6</sup>, -Sphenyl, -SO<sub>2</sub>phenyl, Oalkenyl, or -SiR<sup>6</sup><sub>3</sub> group.

- family of xanthenes of formula (IX):



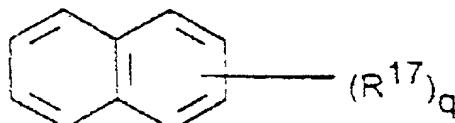
n = 0 to 8

- 10 • family of xanthones of formula (X):



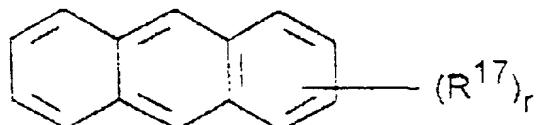
p = 0 to 8

- family of the naphthalene of formula (XI):



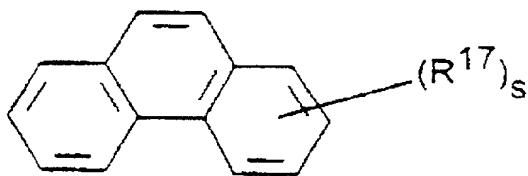
15 q = 0 to 8

- family of the anthracene of formula (XII):



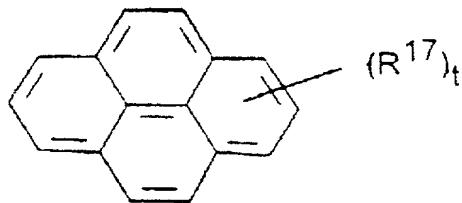
r = 0 to 10

- family of the phenanthrene of formula (XIII):



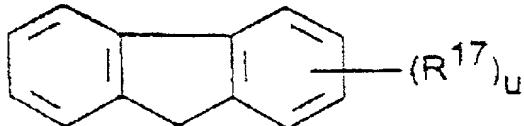
s = 0 to 10

- family of the pyrene of formula (XIV):



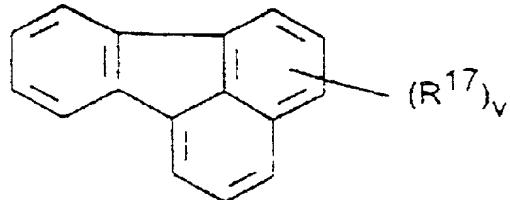
5 t = 0 to 10

- family of the fluorene of formula (XV):



u = 0 to 9

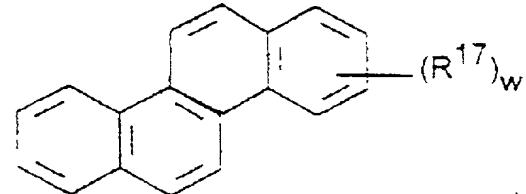
- family of the fluoranthene of formula (XVI):



10

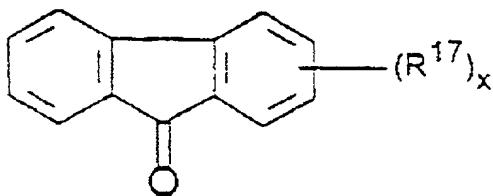
v = 0 to 10

- family of the chrysene of formula (XVII):



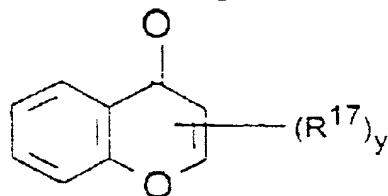
15 w = 0 to 12

- family of the fluorene of formula (XVIII):



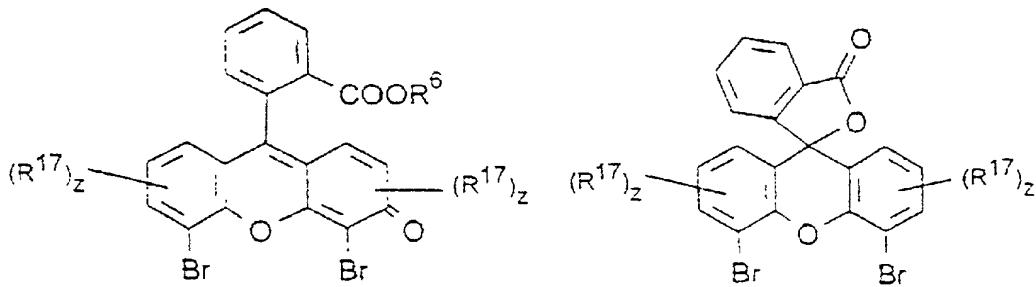
with  $x = 0$  to 8, for example 2,7-dinitro-9-fluorenone,

- family of the chromone of formula (XIX):



with  $y = 0$  to 6

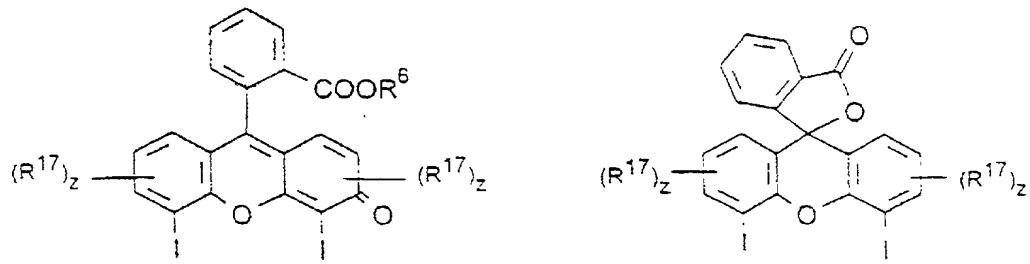
- family of the eosin of formula (XX):



with  $z = 0$  to 5

with  $z = 0$  to 6

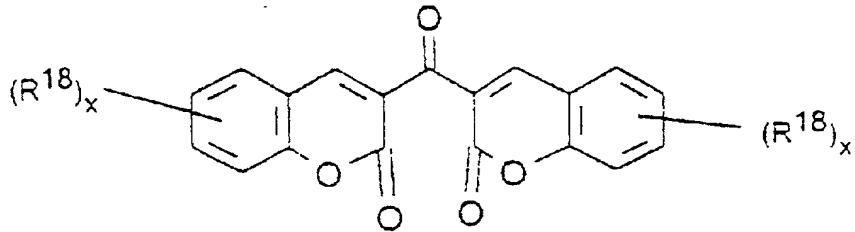
- family of the erythrosin of formula (XXI):



with  $z = 0$  to 5

with  $z = 0$  to 6

- family of the biscoumarins of formula (XXII):



- R<sup>18</sup>, identical or different, has the same meaning as R<sup>17</sup> or represents a group -NR<sup>6</sup><sub>2</sub>, for example 3,3'-carbonylbis(7-diethylaminocoumarin) and 3,3'-carbonyl-5 bis(7-methoxycoumarin).

Other sensitizers can be used. In particular, the photosensitizers described in the documents US 4,939,069; US 4,278,751; US 4,147,552 may be used.

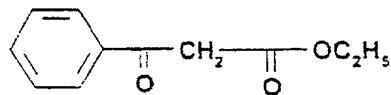
In the context of the present invention, the 10 photosensitizers have a residual absorption of UV light between 200 and 500 nm, preferably 400 to 500 nm for the preparations of dental prostheses. For dental restoration, a photosensitizer having a residual absorption of UV light above 400 nm will be preferred.

15 According to a preferred variant, the photosensitizers will be chosen from those of the families (IV), (VII) and (VIII). By way of examples, the following photosensitizers will be mentioned:

4,4'-dimethoxybenzoin; 2-4-diethylthioxanthone  
2-ethylanthraquinone; 2-methylanthraquinone;  
1,8-dihydroxyanthraquinone; dibenzoylperoxide;  
2,2-dimethoxy-2-phenylacetophenone;  
benzoin;  
2-hydroxy-2-methylpropiophenone;  
benzaldehyde;

4-(2-hydroxyethoxy)phenyl-(2-hydroxy-2-methylpropyl)-  
ketone;

benzoylacetone;



2-isopropylthioxanthone; 1-chloro-4-propoxy-  
thioxanthone;

4-isopropylthioxanthone; and the mixture thereof.

Various types of fillers can be used for preparing the compositions according to the invention. The fillers are chosen according to the final use of  
5 the dental composition: these affect important properties such as appearance, penetration of UV radiation, as well as the mechanical and physical properties of the material obtained after crosslinking and/or polymerization of the dental composition.

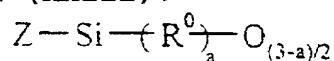
10 As reinforcing filler, there may be used treated or untreated pyrogenic silica fillers, amorphous silica fillers, quartz, glass or nonglassy fillers based on oxides of zirconium, barium, calcium,

fluorine, aluminum, titanium, zinc, borosilicates, aluminosilicates, talc, spherosil, ytterbium trifluoride, fillers based on polymers in ground powder form, such as inert or functionalized methyl 5 polymethacrylates, polyepoxides or polycarbonates.

By way of example, there may be mentioned:

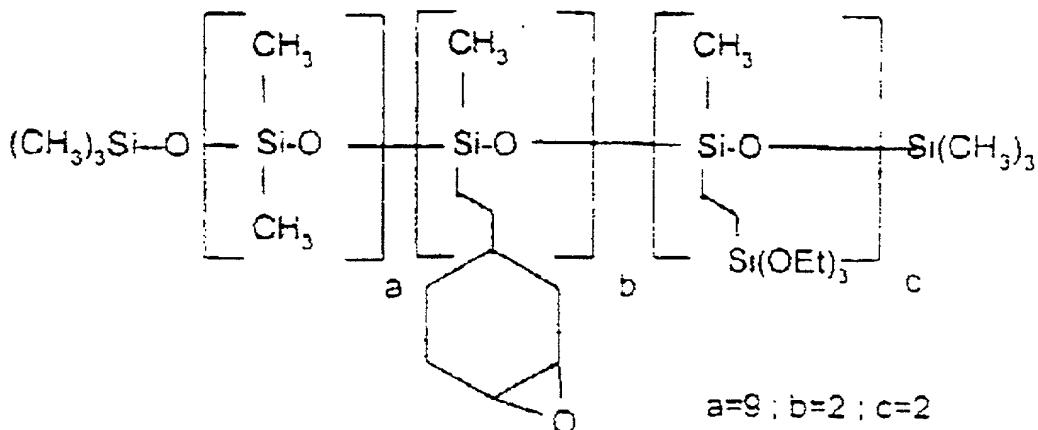
- inert fillers based on methyl polymethacrylate LUXASELF from the company UGL, which can be used in the dental field 10 and which are pigmented in pink,
- hexamethyldisilazane-treated fumed silica fillers having a specific surface area of 200 m<sup>2</sup>/g,
- untreated fumed silica fillers ("aerosil" 15 AE200 marketed by DEGUSSA).

According to an advantageous variant of the invention, the fillers and in particular the silica fillers are treated before use at 120°C with a quantity of less than 10% w/w of silicone comprising at least 20 one unit of formula (XXIII):



- such that Z' has the same definition as Z
- a = 0,1,2 or 3
- with at least one silicon atom.

25 There may be mentioned by way of example the polymer described below with Z = epoxide and Z = trialkoxysilyl



In this case for the treatment of silicone-containing filler(s), in particular silica, with this type of polymer, the material obtained after 5 crosslinking has a mechanical strength, a modulus of elasticity and a resistance to compression which are markedly improved.

In addition to the reinforcing fillers, pigments may be used to color the dental composition 10 according to the invention envisaged and the ethnic groups.

For example, red pigments are used in the presence of microfibers for the dental compositions used for the preparation of dental prostheses in order 15 to simulate the blood vessels.

Pigments based on metal oxides (iron and/or titanium and/or aluminum and/or zirconium oxides, and the like) are also used for the dental compositions used for the preparation of restoration material, in

order to obtain a crosslinked material having an ivory color.

Other additives may be incorporated into the dental compositions according to the invention. For 5 example, biocides, stabilizers, flavoring agents, plasticizers and adherence promoters.

Among the additives which may be envisaged, there will be advantageously used crosslinkable and/or polymerizable coreagents of the organic type. These 10 coreagents are liquid at room temperature or are hot-meltable at a temperature of less than 100°C, and each coreagent comprises at least two reactive functional groups such as oxetane-alkoxy, oxetane-hydroxyl, oxetane-alkoxysilyl, carboxyl-oxetane, oxetane-oxetane, 15 alkenyl ether-hydroxyl, alkenyl ether-alkoxysilyl, epoxy-alkoxy, epoxy-alkoxysilyls, dioxolane-dioxolane-alcohol, and the like.

The dental compositions according to the invention may be used for numerous dental applications, 20 and in particular in the field of dental prostheses, in the field of dental restoration and in the field of temporary teeth.

The dental composition according to the invention is preferably provided in the form of a 25 single product containing the various components ("monocomponent") which facilitates its use, in particular in the field of dental prostheses.

Optionally, the stability of this product may be provided for by organic derivatives with amine functional groups according to the teaching of the document WO 98/07798.

5 In the field of dental prostheses, the product in the "monocomponent" form may be deposited with the aid of a syringe directly on the plaster model or in a core. Next, it is polymerized (polymerization by possible successive layers) with the aid of a  
10 UV lamp (visible light spectrum 200-500 nm).

In general, it is possible to produce a lasting and esthetic dental prosthesis in 10 to 15 min.

It should be noted that the products obtained from the dental composition according to the invention  
15 are nonporous. Thus, after an optional polishing with the aid of a felt brush, for example, the surface of the dental prostheses obtained is smooth and bright and therefore does not require the use of varnish.

The applications in the field of dental  
20 prostheses are essentially those of the joined prosthesis, which can be divided into two types:

- full prosthesis in the case of a patient with absolutely no teeth
- partial prosthesis due to the absence of several  
25 teeth, resulting either in a provisional prosthesis, or a skeleton brace.

In the field of dental restoration, the dental composition according to the invention may be used as material for filling the anterior and posterior teeth in different colors (for example "VITA" colors),  
5 which is rapid and easy to use.

The dental composition being nontoxic and polymerizable in thick layers, it is not essential to polymerize the material in successive layers. In general, a single injection of the dental composition  
10 is sufficient.

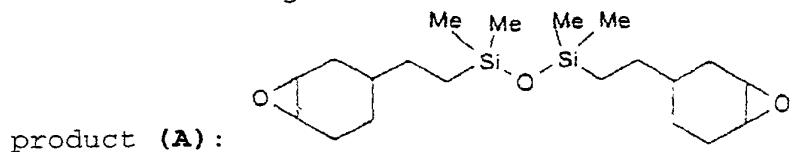
The preparations for dental prostheses and for restoration materials are carried out according to techniques which are customary in the art.

In the case of application of the dental  
15 composition as a tooth, either the tooth may be pretreated with a bonding primer or the dental composition may be prepared as a mixture with a bonding primer before its use. However, it is not essential to use a bonding primer in order to use the dental  
20 composition according to the invention.

The following examples and tests are given by way of illustration. They make it possible in particular to understand more clearly the invention and to highlight some of its advantages and to illustrate a  
25 few of its variant embodiments.

Examples and tests

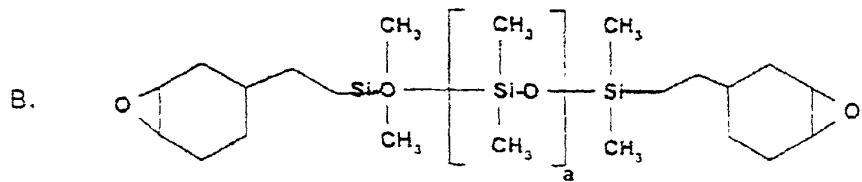
The product used in the compositions of the examples are the following:



5

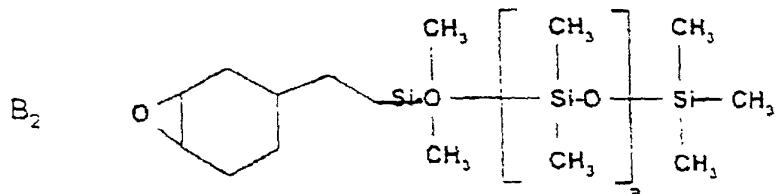
Product (B): this product is a mixture of siloxanes, whose viscosity is 23.5 mPa.s and in which the proportions by weight and formulae B<sub>1</sub>, B<sub>2</sub> and B<sub>3</sub> are given below:

10

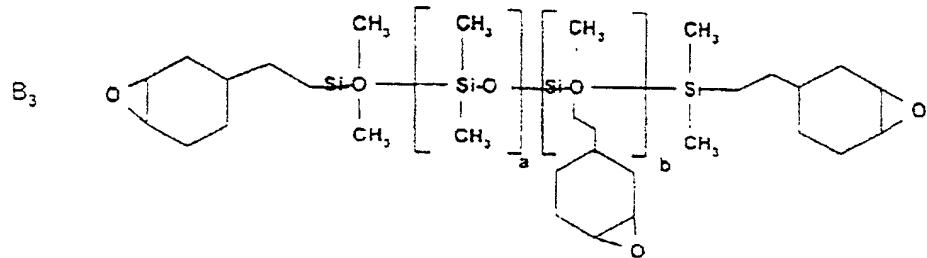


with 89% of B<sub>1</sub> where  $a = 0$ , 9% of B<sub>1</sub> where  $a = 1$ ; 0.2% of B<sub>1</sub> where  $a = 2$ ;

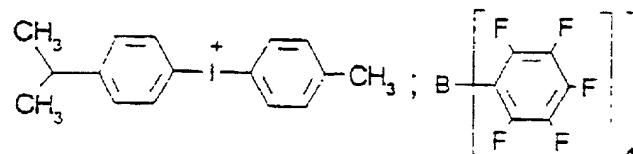
15



with 0.3% of B<sub>2</sub> where  $a = 0$ ;



and with 1.5% of B3 where  $a = 0$  and  $b = 1$ .



5 product (P1) :

Product (**PS1**): isopropylthioxanthone marketed under the trademark Quantacure ITX by the company RAHN.

## Example 1 - Composition for dental prostheses

The following are mixed using a three-blade stirrer:

- 100 parts of siloxane (**A**) stabilized with 50 ppm of Tinuvin 765;
  - 1 part of photoinitiator (**P1**) at 75% in ethyl acetate;
  - 0.028 part of photosensitizer (**PS1**);
  - 150 parts of a pink-pigmented, polymethyl methacrylate-based inert filler (product LUXASELF from UGL dentaire).

The composition obtained is perfectly stable in the absence of light for several months at room temperature. This composition can be worked manually and for several hours in the presence of daylight.

- 5 A test piece  $2.8 \pm 0.3$  mm thick is prepared in a glass dish 64 mm long (model), 10 mm wide (model) and open at the top by pouring the prepared composition ("monocomponent") into the dish.

- 10 The composition is dried by passing the dish for 1 to 2 seconds (3 m/min) under a UV lamp of 200 W/cm power corresponding to the excitation of a mixture of mercury and gallium and emitting in the UV-visible range above 400 nm.

- 15 The product obtained is unmolded by breaking the glass.

The SHORE D hardness of the two polymerized compositions is determined on each side of the item made immediately after crosslinking.

Example 1	Immediate measurement	Measurement after 10 hours
Irradiated surface:	70	85
Bottom surface	60	85

- 20 The Shore D hardness continues to change substantially over a few hours.

The volume shrinkage is very low and excellent size stability is obtained.

The loss of mass is less than 1%.

- 5 The product may be used with or without bonding primer in the presence of artificial teeth or of natural teeth.

More generally, the properties of the material obtained are in agreement with the DIN/ISO 1567 standard.

10 **Example 2 - Composition for dental restoration**

A composition for dental restoration is prepared by mixing:

- 15 - 200 parts of siloxane (**A**) stabilized with 50 ppm of Tinuvin 765;
- 1.8 parts of photoinitiator (**P1**) at 75% in ethyl acetate;
- 0.0178 part of photosensitizer (**PS1**);
- 52 parts of hexamethyldisilazane-treated fumed silica having a specific surface area of 200 m<sup>2</sup>/g;
- 20 - 20 parts of untreated amorphous silica dried for 4 hours at 200°C before formulation.

A flowing composition is obtained which has a translucent gray appearance.

The crosslinking-polymerization operation is carried out using a lamp emitting a light spot emitted

through a curved light tip 8 mm in diameter. The source is an Optibulb 80 W lamp (DEMETRON Optilux 500) for wavelengths of between 400 and 520 nm.

The dental composition is applied in a tooth.

- 5 A thickness of 5 mm is crosslinked in less than 30 seconds.

A SHORE D hardness of 50 is obtained immediately and can reach 80 to 100 within a few hours.

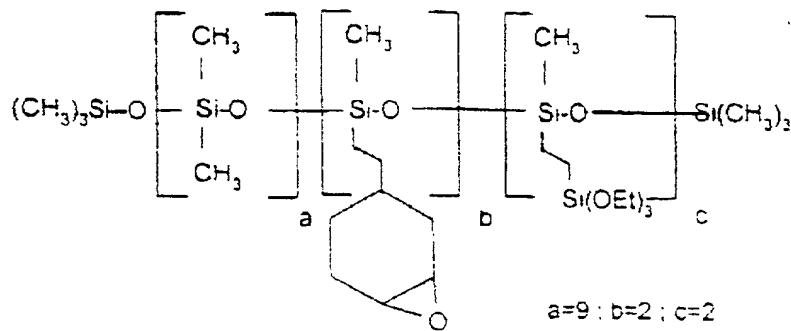
- No loss is observed in size stability. The  
10 porosity of the crosslinked material, based on observation of a section under a microscope, is zero.

The color of the composite after crosslinking is close to ivory color.

**Example 3 - Composition for dental restoration**

- 15 The same concentrations of components and the same procedure as above in Example 2 are used.

However, the mixture of fillers is treated, before use, at 120°C with 5% w/w of silicone of average general formula containing less than 50 ppm of residual  
20 platinum:



The restoration material obtained after crosslinking according to the procedure of Example 2 has a better mechanical strength, an improved modulus 5 of elasticity and a better compression resistance.

A SHORE D hardness of 80 is obtained immediately.

The size stability and the porosity properties of the material are excellent.

10 **Example 4 - Dental precomposition**

A dental precomposition, prepared without fillers, is obtained by mixing using a magnetic stirrer bar:

- 15           - 1 part of siloxane (**A**) having a density of 0.997, stabilized with 50 ppm of Tinuvin 765,  
          - 0.01 part of photoinitiator (**P1**) at 10% in solution, dissolved directly in siloxane A,  
          - 0.00028 part of photosensitizer (**PS1**) contained in the photoinitiator (**P1**).

20           The crosslinking operation is carried out in a manner identical to that of Example 2. A composition 5 mm thick is crosslinked in less than 30 seconds.

The density of the crosslinked composition is measured using a brass pycnometer and its value is 25 1.0274. The volume shrinkage is therefore 3.05% [ $= (1.0274 - 0.997) / 0.997 \times 100$ ] in the absence of fillers. Consequently, a dental composition of this type,

formulated with more than 50% of filler, will have a very low volume shrinkage which is less than 1.5%.

Example 5 - Dental precomposition

(a) A dental precomposition prepared without  
5 fillers is obtained by mixing using a three-blade  
stirrer,

- 10 parts of siloxane (**A**) having a density  
of 0.997, stabilized with 50 ppm of Tinuvin  
765,

10 - 0.01 part of photoinitiator (**P1**) dissolved  
directly in silicone (A),  
- and 62 ppm of photosensitizer (PS1)  
contained in the photoinitiator.

Eight grams of this dental precomposition are  
15 then placed in an open cylindrical aluminum cup in a  
manner such that the volume occupied represents a  
thickness of about 6 mm.

The liquid is photocrosslinked by passing the  
cup under a UV lamp delivering, through a pane of glass  
20 6 mm thick, UV-V (> 390 nm) = 0.6 W/cm<sup>2</sup>. The  
UV-V (> 390 nm) dose received is 0.4 J/cm<sup>2</sup> at 10 m/min.

The degree of conversion of the epoxy  
functional groups immediately after polymerization is  
measured from the residual heat of reaction recorded by  
25 differential calorimetry. The residual heat is 17 J/g  
relative to a noncrosslinked sample which represents  
237 J/g. The effective degree of conversion of the

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epoxy functional groups with 0.1% photoinitiator is therefore 93%.

- (b) The same composition as above in (a) is prepared by adding 5% w/w of 3-ethyl-  
5 3-(hydroxymethyl)oxetane during the preparation of the composition. The composition is then crosslinked in the same manner as above in (a).

The degree of conversion of the epoxy functional groups is 99.3% with a residual heat of  
10 1.5 J/g.

- (c) The same composition as above in (a) is prepared by adding 10% w/w of 3-ethyl-  
3-(hydroxymethyl)oxetane during the preparation of the composition. The composition is then crosslinked in the same manner as above in (a).  
15

The degree of conversion of the epoxy functional groups is 99.95% with a residual heat of 0.1 J/g.

Example 6 - Composition for dental prosthesis or dental restoration material

The following are mixed using a three-blade stirrer:

- 95 parts of siloxane (**A**) stabilized with 50 ppm of Tinuvin 765,
- 25 62 ppm of photosensitizer (**PS1**),
- 0.5 part of photoinitiator (**P1**) at 10% in siloxane (**A**),

- 5 parts of 3-ethyl-3-(hydroxymethyl)-oxetane,

- and 120 parts of precipitated silica (ground quartz).

5 A dental composition is obtained which is opaque, gray in color, nonflowing and easy to handle.

The crosslinking operation is carried out in a manner identical to that of Example 2. A composition 5 mm thick crosslinks in less than 30 seconds.

10 The color of the material after crosslinking is close to ivory color.

The material is suitable in particular for dental prosthesis use, in particular the stiffness is greater than 80 Mpa according to the ISO 1567 standard.

15 **Example 7 - Dental composition**

This composition is formulated with:

- 95 parts of silicone (B),
- 62 ppm of photosensitizer (PS1),
- 0.5 part of photoinitiator (P1) at 10% in siloxane (B),
- 5 parts of the oxetane 3-ethyl-3-(hydroxymethyl)oxetane,
- and 120 parts of precipitated silica (ground quartz).

25 The crosslinking operation is carried out in a manner identical to that of Example 2.

The stiffness values found are greater than 80 Mpa according to the ISO 1567 standard.

**Example 8 - Dental precomposition**

A dental precomposition **M1** prepared without  
5 fillers is obtained by mixing using a three-blade  
stirrer:

- 100 parts of a silicone composition (**B**)

having a density of 0.997 and stabilized with  
50 ppm of Tinuvin 765,

10 - 0.1 part of photoinitiator (P1) at 10% in  
solution, dissolved directly in silicone (B),  
- and 0.028 part of photosensitizer (**PS1**).

A portion of the composition obtained **M1** is removed so as to crosslink it. The crosslinking  
15 operation is carried out in a manner identical to that of Example 2. A composition 5 mm thick crosslinks in less than 30 seconds.

The density of the crosslinked material, measured using a brass pycnometer, is 1.0274. The  
20 volume shrinkage is therefore 3.05%  
 $[=(1.0274-0.997)/0.997 \times 100]$  in the absence of fillers. Consequently, a dental composition of this type, formulated with more than 50% of fillers, will have a very low volume shrinkage which is less than 1%.

Example 9 - Dental precomposition

(a) Eight grams of **M1** of Example 8 are placed in an open cylindrical aluminum cup such that the volume occupied represents a thickness of about 6 mm.

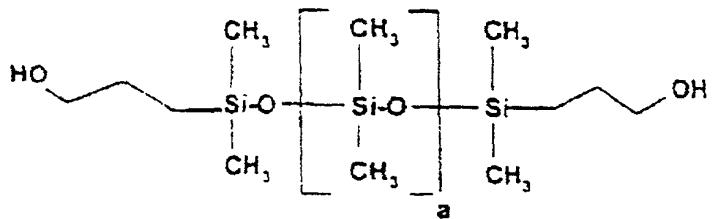
5       The photocrosslinking of **M1** and the calculation of the degree of conversion of the epoxy functional groups are carried out according to the methods described in Example 5.

The residual heat is 17 J/g relative to a  
10 noncrosslinked sample which represents 237 J/g. Consequently, the effective degree of conversion of the epoxy functional groups with 0.1% of photoinitiator is therefore 93%.

(b) The photocrosslinking and the calculation  
15 of the degree of conversion of the epoxy functional groups are also carried out for a composition **M1** containing 5% w/w of a silicone oil of formula  $B_1$  with  $a = 12$  on average.

The degree of conversion of the epoxy  
20 functional groups is 99% with a residual heat of 1.5 J/g.

(c) The photocrosslinking and the calculation  
of the degree of conversion of the epoxy functional  
groups are also carried out for a composition **M1**  
25 containing 5% w/w of a silicone oil with  $a = 12$  on average and of formula:



The degree of conversion of the epoxy functional groups is 99% with a residual heat of  
5 0.1 J/g.

(d) The photocrosslinking and the calculation of the degree of conversion of the epoxy functional groups are also carried out for a composition **M1** containing 5% by weight of siloxane resin of the MQM<sup>Epoxy</sup> type containing 0.5% by weight of hydroxysilyl functional groups and 2% by weight of epoxy functional group (mass 43).

The degree of conversion of the epoxy functional groups is 99%.

15 **Example 10 - Composition for dental prosthesis or dental restoration material**

The following are mixed using a three-blade stirrer:

- 100 parts of silicone (**B**),
- 20 - 62 ppm of photosensitizer (**PS1**),
- 0.5 part of photoinitiator (**P1**) at 10% in solution in silicone (**B**),
- and 120 parts of precipitated silica (ground quartz).

A mixture is obtained which is opaque, gray in color, nonflowing and capable of being handled.

The crosslinking operation is carried out in a manner identical to that of Example 2. A composition  
5 5 mm thick is crosslinked in less than 30 seconds. The color of the material after crosslinking is similar to ivory color.

The composition, in this case, is suitable in particular for dental prostheses, in particular the  
10 stiffness is greater than 80 Mpa according to the ISO 1567 standard.

**Example 11 - Dental composition**

The following are mixed using a three-blade stirrer:

- 15           - 95 parts of silicone (**B**) stabilized with  
              50 ppm of Tinuvin 765,  
              - 62 ppm of photosensitizer (**PS1**),  
              - 0.5 part of photoinitiator (**P1**) at 10% in  
              silicone (**B**),  
20           - 5 parts of polydimethylsiloxane **B1** where a  
              = 12 on average,  
              - and 120 parts of precipitated silica  
              (ground quartz).

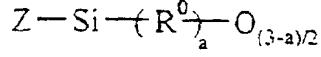
The crosslinking operation is carried out in  
25 a manner identical to that of Example 2. The measured stiffness values are greater than 80 Mpa according to the ISO 1567 standard.

CLAIMS

1. A dental composition comprising:

(1) at least one crosslinkable and/or polymerizable silicone oligomer or polymer which is liquid at room temperature or which is heat-melttable at a temperature of less than 100°C, and which comprises:

- at least one unit of formula (FS):



10

in which:

15

- a = 0, 1 or 2,
- $\text{R}^0$ , identical or different, represents an alkyl, cycloalkyl, aryl, vinyl, hydroxyl or alkoxy radical, preferably a  $\text{C}_1\text{-}\text{C}_6$  lower alkyl,

20

- Z, identical or different, is an organic substituent comprising at least one reactive epoxy, and/or alkenyl ether and/or oxetane and/or dioxolane and/or carbonate functional group,

- and at least two silicon atoms,

25

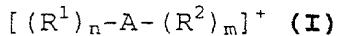
- (2) at least one aromatic hydrocarbon photosensitizer with one or more aromatic nuclei which are substituted or not, having a residual light absorption of between 200 and 500 nm,

(3) at least one dental filler present in a proportion of at least 10% by weight relative to the total weight of the composition,

5 (4) and an effective quantity of at least one borate-type photoinitiator, chosen from those of formula:

Δ in which the cationic entity of the borate is selected from:

(\*) the onium salts of formula (I):



10 in which formula:

• A represents an element of groups 15 to 17 such as for example: I, S, Se, P or N,

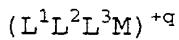
15 • R<sup>1</sup> represents a carbocyclic or heterocyclic C<sub>6</sub>-C<sub>20</sub> aryl radical, it being possible for said heterocyclic radical to contain, as heteroelements, nitrogen or sulfur,

20 • R<sup>2</sup> represents R<sup>1</sup> or a linear or branched C<sub>1</sub>-C<sub>30</sub> alkyl or alkenyl radical; said radicals R<sup>1</sup> and R<sup>2</sup> being optionally substituted with a C<sub>1</sub>-C<sub>25</sub> alkoxy, C<sub>1</sub>-C<sub>25</sub> alkyl, nitro, chloro, bromo, cyano, carboxyl, ester or mercapto group,

• n is an integer ranging from 1 to v + 1, v being the valency of the element A,

25 • m is an integer ranging from 0 to v - 1 with n + m = v + 1,

(\*\*) the organometallic salts of formula (III):



in which formula:

- M represents a group 4 to 10 metal, in particular iron, manganese, chromium or cobalt,
- 5     • L<sup>1</sup> represents 1 ligand bound to the metal M by  $\pi$  by electrons, which ligand is chosen from the ligands  $\eta^3$ -alkyl,  $\eta^5$ -cyclopentadienyl and  $\eta^7$ -cycloheptatrienyl and the  $\eta^6$ -aromatic compounds chosen from the optionally substituted  $\eta^6$ -benzene ligands and the compounds having from 2 to 4 condensed rings, each ring being capable of contributing to the valency layer of the metal M by 3 to 8  $\pi$  electrons;
- 10    • L<sup>2</sup> represents a ligand bound to the metal M by  $\pi$  electrons, which ligand is chosen from the ligands  $\eta^7$ -cycloheptatrienyl and the  $\eta^6$ -aromatic compounds chosen from the optionally substituted ligands  $\eta^6$ -benzene and the compounds having from 2 to 4 condensed rings, each ring being capable of contributing to the valency layer of the metal M by 6 or 7  $\pi$  electrons;
- 15    • L<sup>3</sup> represents from 0 to 3 ligands, which are identical or different, linked to the metal M by  $\sigma$  electrons, which ligand(s) is (are) chosen from CO and NO<sub>2</sub><sup>+</sup>; the total electron charge q of the complex to which L<sup>1</sup>, L<sup>2</sup> and L<sup>3</sup> contribute and the

ionic charge of the metal M being positive and equal to 1 or 2;

Δ the anionic borate entity of which has the formula  $[BX_aR_b]^-$  in which:

5 - a and b are integers ranging, for a, from 0 to 3 and, for b, from 1 to 4 with  $a + b = 4$ ,

- the symbols X represent:

\* a halogen atom (chlorine, fluorine) with a = 0 to 3,

10 \* an OH functional group with a = 0 to 2,

- the symbols R are identical or different and represent:

15 ➤ a phenyl radical substituted with at least one electron-attracting group such as for example OCF<sub>3</sub>, CF<sub>3</sub>, NO<sub>2</sub>, CN, and/or with at least 2 halogen atoms (fluorine most particularly), this being when the cationic entity is an onium of an element of groups 15 to 17,

20 ➤ a phenyl radical substituted with at least one element or one electron-attracting group, in particular a halogen atom (fluorine most particularly), CF<sub>3</sub>, OCF<sub>3</sub>, NO<sub>2</sub>, CN, this being when the cationic entity is an organometallic complex of an element of groups 4 to 10,

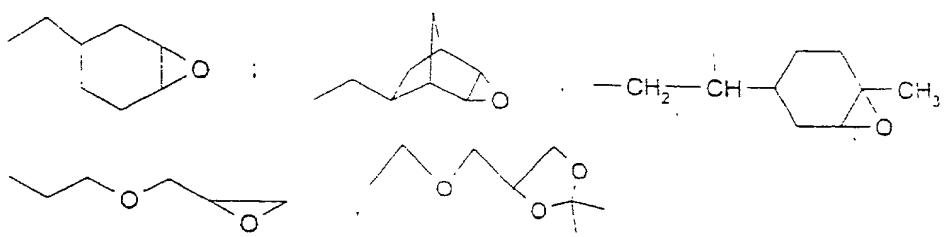
25 ➤ an aryl radical containing at least two aromatic nuclei such as for example biphenyl, naphthyl, optionally substituted with at least one electron-

attracting group or element, in particular a halogen atom, including fluorine in particular, OCF<sub>3</sub>, CF<sub>3</sub>, NO<sub>2</sub>, CN, regardless of the cationic entity.

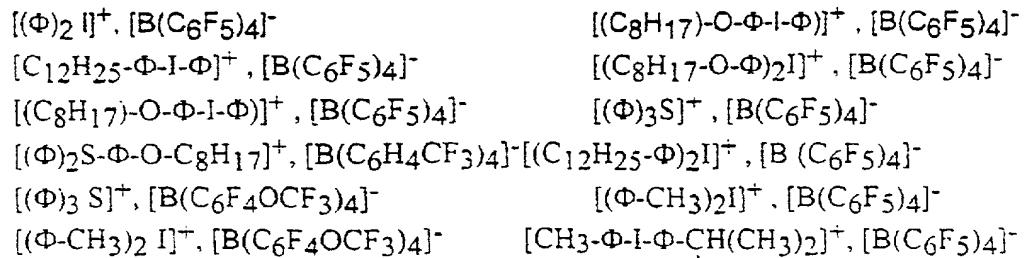
5           2. The composition as claimed in claim 1, characterized in that Z is an organic substituent Z1 comprising at least one reactive epoxy, and/or dioxolane functional group, and preferably at least one reactive epoxy functional group.

10          3. The composition as claimed in claim 2, characterized in that the oligomer or polymer (1) comprises in addition other reactive functional groups Z such as the reactive alkenyl ether, oxetane and/or carbonate functional groups Z2.

15          4. The composition as claimed in any one of the preceding claims, characterized in that the reactive functional group(s) of Z1 are chosen from the following radicals:



5. The composition as claimed in any one of the preceding claims, characterized in that the photoinitiator is chosen from the group consisting of:

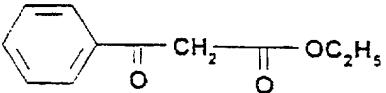


- 5            $(\eta^5\text{-cyclopentadienyl})(\eta^6\text{-toluene}) Fe^+$ ,  $[B(C_6F_5)_4]^-$   
              $(\eta^5\text{-cyclopentadienyl})(\eta^6\text{-methyl-1-naphthalene}) Fe^+$ ,  
              $[B(C_6F_5)_4]^-$   
              $(\eta^5\text{-cyclopentadienyl})(\eta^6\text{-cumene}) Fe^+$ ,  $[B(C_6F_5)_4]^-$

and the mixture thereof.

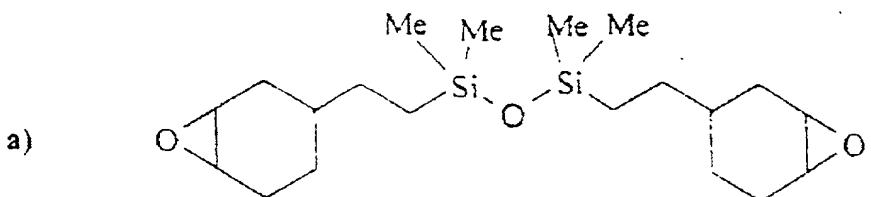
10          6. The composition as claimed in any one of the preceding claims, characterized in that the photosensitizer is chosen from the group consisting of:

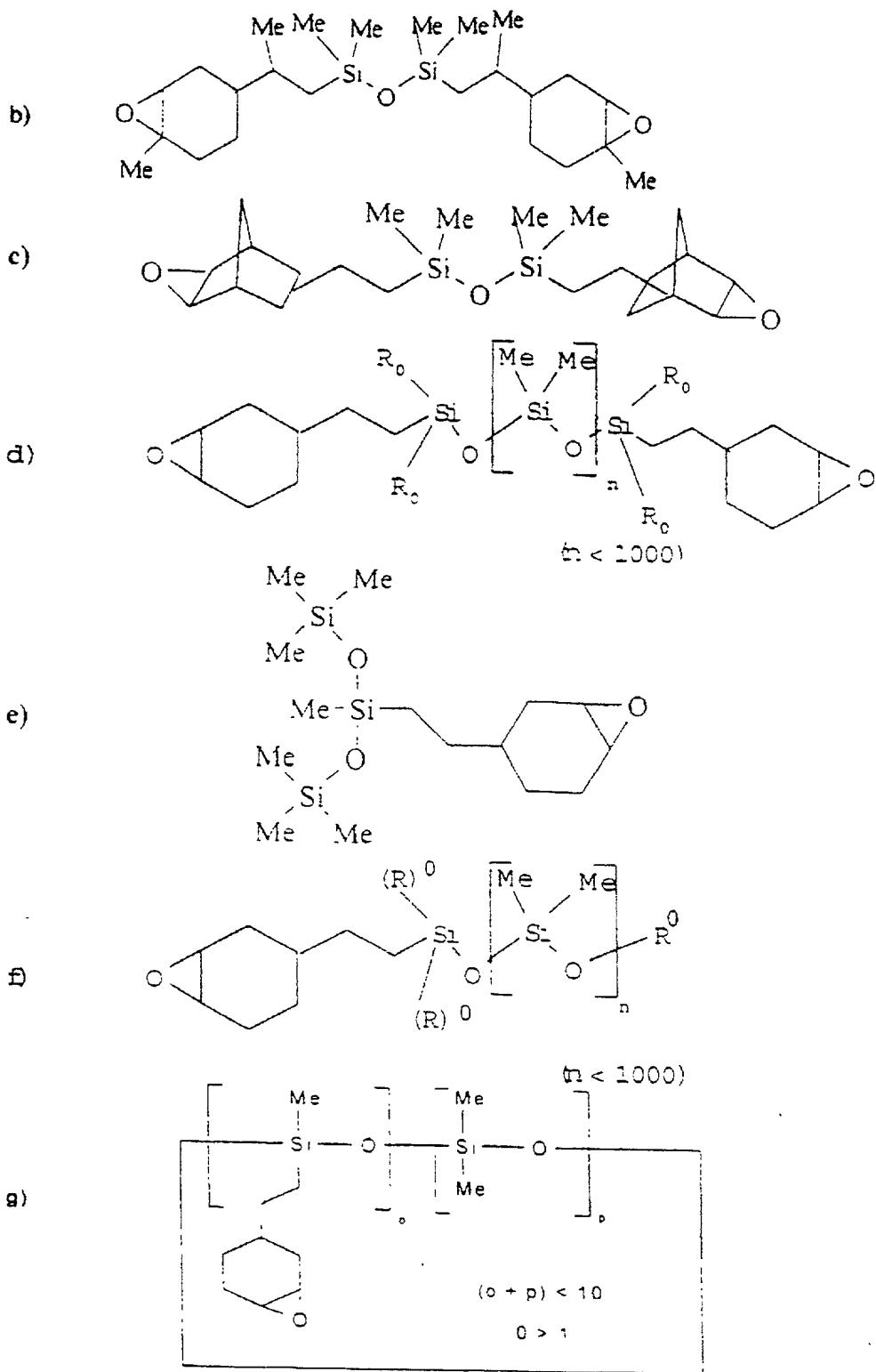
4, 4'-dimethoxybenzoin;                    2-4-diethylthioxanthone  
2-ethylanthraquinone;                        2-methylanthraquinone;  
1, 8-dihydroxyanthraquinone;                dibenzoylperoxide;  
2, 2-dimethoxy-2-phenylacetophenone;  
    benzoin;  
2-hydroxy-2-methylpropiophenone;  
    benzaldehyde;  
4-(2-hydroxyethoxy)phenyl-(2-hydroxy-2-methylpropyl)-  
ketone;  
benzoylacetone;

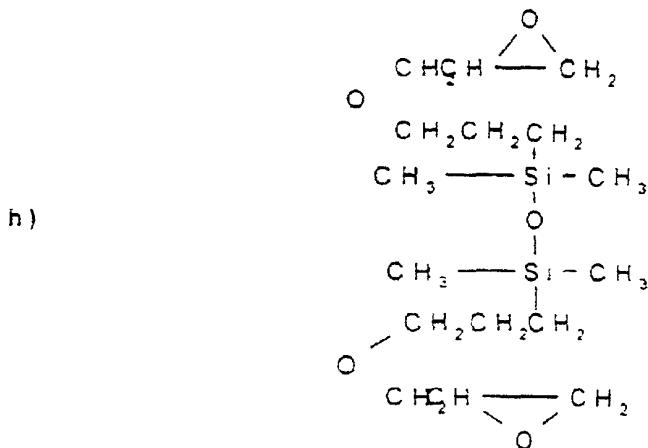


2-isopropylthioxanthone;                    1-chloro-4-propoxy-  
    thioxanthone;  
4-isopropylthioxanthone;                    and the mixture thereof.

7. The dental composition as claimed in any one of the preceding claims, characterized in that the silicone oligomer and/or polymer (1) consists of at least one silicone having the following average formula:







8. The use of a dental composition as  
5 claimed in any one of the preceding claims for the  
production of dental prostheses.

9. The use of a dental composition as  
claimed in any one of claims 1 to 7, for dental  
restoration.

10 10. A dental prosthesis which can be  
obtained from a composition as claimed in any one of  
claims 1 to 7.

11. A dental restoration material which can  
be obtained from a composition as claimed in any one of  
15 claims 1 to 7.

**DENTAL COMPOSITION BASED ON SILICONE CROSSLINKABLE BY  
CATION PROCESS**

5

**Abstract of the Disclosure**

The invention concerns dental compositions. Said composition comprises (1) a silicone crosslinkable and/or polymerizable by cation process; (2) an efficient amount of at least an initiator such as onium borate; (3) at least a photosensitizer ; and (4) a dental filler present in the composition in a proportion of at least 10 wt. % relative to the 10 composition total weight. Said dental compositions are useful for making dental prostheses or for dental restoration.

15 20 20 C/Data/RN98131.Abst

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COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY  
(include Reference to PCT International Applications) PCT/FR99/02345

ATTORNEY'S DOCKET NO  
**RN98131**

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

**DENTAL COMPOSITION BASED ON SILICONE CROSSLINKABLE BY CATION PROCESS**

the specification of which (check only one item below):

- is attached hereto.  
 was filed as United States application

Serial No. \_\_\_\_\_

on \_\_\_\_\_

and was amended

on \_\_\_\_\_ (if applicable)

- was filed as PCT international application

Number **PCT/ FR99/02345**

on **October 1st, 1999**

and amended under PCT ARTICLE 19

on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the patentability of this application in accordance with Title 37, Code of Federal Regulations. §1.56(a).

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed:

**PRIOR FOREIGN/PCT APPLICATION(S) AND ANY PRIORITY CLAIMS UNDER 35 U.S.C. 119:**

COUNTRY PCT indicate PCT	APPLICATION NUMBER	DATE OF FILING (day month year)	PRIORITY CLAIMED UNDER 35 USC 119	
FRANCE	98/12375	02 October, 1998	<input checked="" type="checkbox"/> YES	<input type="checkbox"/> NO
			<input type="checkbox"/> YES	<input type="checkbox"/> NO
			<input type="checkbox"/> YES	<input type="checkbox"/> NO
			<input type="checkbox"/> YES	<input type="checkbox"/> NO
			<input type="checkbox"/> YES	<input type="checkbox"/> NO

COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY (Continued) (include Reference to PCT International Applications) PCT/FR99/02345		ATTORNEY'S DOCKET NO <b>RN98131</b>																									
<p>I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) or PCT international application(s) designating the United States of America that is/are listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in that those prior application(s) in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application(s) and the national or PCT international filing date of this application:</p>																											
<b>PRIOR U.S. APPLICATIONS OR PCT INTERNATIONAL APPLICATIONS DESIGNATING THE U.S. FOR BENEFIT UNDER 35 U.S.C. 120</b>																											
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<p><b>POWER OF ATTORNEY:</b> As a named inventor, I hereby appoint the following attorney's and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith (List name and registration number) JOHN A. SHEDDEN - Reg. No. <u>25,644</u>,  <span style="margin-left: 40px;">3</span> Kevin McVEIGH - Reg. No. <u>33,017</u>          JOHN D. WOOD - Reg. No. <u>31,146</u>          JEAN-LOUIS SEUGNET - Limited Recognition under 37 CFR § 10.9(b) enclosed</p>																											
<p>Send Correspondence to:  <u>JEAN-LOUIS SEUGNET</u>  <u>INTELLECTUAL PROPERTY DEPT.</u>  <u>RHODIA INC.</u>  <u>259 PROSPECT PLAINS ROAD,</u>  <u>CN 7500, CRANBURY, NJ 08512-7500</u></p>		<p>Direct Telephone Calls to:          (name and telephone number)  <u>JEAN-LOUIS SEUGNET</u>  <u>(609) 860-4180</u></p>																									
<b>201</b>	FULL NAME OF INVENTOR	FAMILY NAME	FIRST GIVEN																								
		<u>FRANCES</u>	<u>Jean-Marc</u>																								
	RESIDENCE & CITIZENSHIP	CITY	STATE OR FOREIGN COUNTRY																								
		<u>MEYZIEU</u>	<u>FRANCE</u>																								
	POST OFFICE ADDRESS	POST OFFICE ADDRESS	CITY																								
		<u>1, rue de Flandres</u>	<u>MEYZIEU</u>																								
			STATE & ZIP CODE/COUNTRY																								
			<u>F-69330, FRANCE</u>																								
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	POST OFFICE ADDRESS	CITY	STATE & ZIP CODE/COUNTRY																								
<p>I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code; and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.</p>																											
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DATE <u>May 12th, 2001</u>		SIGNATURE OF INVENTOR 203																									
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